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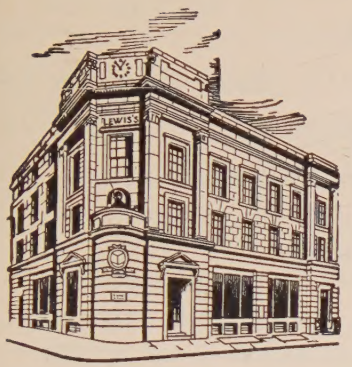

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THE PROCEEDINGS OF THE PHYSICAL SOCIETY

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FURTHER EXPERIMENTS ON THE PHOTOGRAPHIC ACTION OF GAMMA RAYS AND ON THEIR ABSORPTION COEFFICIENTS

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ABSTRACT. The photographic action of γ rays is intensified by using technical X-ray intensifying screens more than by using lead foil. These intensifying screens, however, intensify the longer wave-lengths more than the shorter. It has been found that the heterochromatic γ-ray spectrum from radium (B+C) can be analysed into two apparently homogeneous components. Both components, however, appear to have an effective wave-length which is shorter the higher the atomic number of the absorbing material. These results can be explained, at least approximately, by a consideration of the absorption of the main lines in the γ-ray spectrum of radium (B+C). This explanation suggests the presence in this spectrum of lines of moderate intensity of wave-length less than 5·6 X.

§ 1. INTENSIFYING-SCREENS FOR γ RAYS

IN two earlier papers[1,2] an account has been given of the use of the photographic action of γ rays in the determination of the absorption coefficients of the γ rays from radium (B + C) in a number of elements. In these experiments the photographic action was intensified by using thin lead foil on either side of the Agfa duplitized X-ray film, the lead foil increasing the photographic action approximately 1·7 times.

Frilley[3] has mentioned, without giving quantitative results, that the best intensifying screens for γ-ray photography were those used for X-ray diagnosis. It was decided to investigate these. The type of screen used was the Patterson Speed Intensifying Screen of the thin type, which has a highly glossy surface. It was found that such screens used in conjunction with either Agfa or Ilford X-ray duplitized film produced a fairly uniform photographic blackening but not as uniform as that produced when lead intensifying screens were employed.

In these tests an aluminium film holder was made so that the film could be placed (1) between two pieces of aluminium or (2) between two pieces of lead foil or (3) between two pieces of the Patterson Screen. The film was placed, for each intensifier, at the same distance from the source, and distance pieces of aluminium sheet were used on the side of the film distant from the source to ensure that the film should be tightly packed in the film holder. The intensifying action was examined for γ rays from radium (B + C) under two conditions of filtering. In the first a single glass tube containing 100 mg. of radium was employed so that the rays

were lightly filtered, partly in the glass wall of the tube and partly by the aluminium of the film holder between source and film. The total filtering amounted approximately to 6 mm. of aluminium. In the second 400 mg. of radium in 4 glass tubes in line was filtered by 1·42 cm. of lead so that the total filtering amounted to the equivalent of 1·6 cm. of lead. With this filtering it has been already shown[3] that the γ rays from radium (B + C) are exponentially absorbed.

The time of exposure was chosen for each method of intensification so that the photographic density was less than 0·8, i.e., in the range of densities for which the density is proportional to the time of exposure. In order that all the films, which were to form one set in a comparison, should be developed under identical conditions they are all developed together and fixed together. The measured photographic actions are set out in table 1 below, in which the photographic action produced with aluminium on both sides of the film is taken as unity.

Table 1.* Relative intensifying action for γ rays. Agfa film

Intensifier	Relative photographic action	
	Filter 6 mm. aluminium	Filter 1·6 cm. lead
Aluminium	1	1
Lead foil	1·70 ± 0·04	1·80 ± 0·06
Patterson screen	5·88 ± 0·02	3·65 ± 0·02
Ilford film		
Aluminium	1	1
Lead foil	1·66 ± 0·04	1·54 ± 0·02
Patterson screen	6·1	3·9

One interesting point about the results is that there is very little variation in the ratio of the intensifying action of lead to aluminium with the change of filtering, in other words, with the change in effective wave-length of the γ rays. This holds for both types of films examined. On the other hand, when Patterson screens are used, the intensifying action is much greater for lightly filtered radiations, having the longer effective wave-lengths, than for the more heavily filtered radiation.

Thus if a mixed beam of γ rays consisting, say, of a hard and a soft component is to be photographically analysed by absorption in such a way that the relative intensities of the two components are to be measured, it is necessary to use lead intensifying screens. On the other hand, if it is a question of determining an absorption coefficient, then there is no objection to using the X-ray screens provided it can be assumed that the effective wave-length of the beam remains constant as soon as the absorption becomes exponential. When it was decided to measure absorption coefficients under different filterings, in the manner described below under § 4, Patterson X-ray screens were used, as these considerably reduced the time of exposure. It was, however, first necessary to show that the variation of

* A comparison was also made of the photographic action produced in Agfa and Ilford films exposed under the same conditions. It was found for a filter of 6 mm. of aluminium that the photographic density produced in Ilford was twice that produced in Agfa film, and with a filter of 1·6 cm. of lead the ratio of densities Ilford : Agfa = 1·6.

photographic density with time of exposure, for constant intensity of source, was linear over a range of densities. For both Agfa and Ilford films this law was found to hold over the range of densities 0·3 to 0·8. It was necessary also to investigate the variation of density with intensity, and although this was not done directly, it was done by finding, with the films intensified by X-ray screens, the absorption coefficients in lead of the γ rays from radium (B + C) filtered by 1·6 cm. of lead. The results are set out in table 2, in which it will be seen that the same values of the absorption coefficient are obtained, within the limits of experimental error, whether aluminium, lead or Patterson screens are used to intensify the photographic action.

Table 2. Values of the absorption coefficient in lead

(Filter 1·6 cm. of lead)

Intensifying screen and film	Value of ($cm.^{-1}$)
Aluminium or lead with Agfa	$0{\cdot}532 \pm 0{\cdot}004$
Patterson screen with Agfa	$0{\cdot}539 \pm 0{\cdot}008$
Patterson screen with Ilford	$0{\cdot}523 \pm 0{\cdot}005$

§ 2. THE DEVELOPMENT OF FILMS

In the previous experiments with Agfa film a metol-hydroquinone developer was used, the development taking 5 minutes at 18° C. In any experiment in which different films had to be compared they were developed together under constant agitation to ensure that they should receive the same conditions of development. When, owing to the greater photographic action produced in Ilford film, its use was decided upon, the variation in density produced by varying conditions of development was investigated. A number of pieces of film were exposed under identical conditions to γ rays and these were developed in pairs, together with a piece of unexposed film. From the last-named the density of the chemical fog was measured. The variation of density with time of development (3 to 6 minutes) at 18° C. is shown by curve *A*, figure 1. The variation of density with temperature of developer, for 5 minutes' development, is shown by curves *B* and *C* in the same figure. Curve *B* was obtained with a freshly mixed sample of developer and *C* with the same developer after it had been stored in a dark bottle for 3 days. It will be seen from the curves that if the chemical fog is to be kept below a reasonable amount the temperature of development must not exceed 20° C. for 5 minutes' development. In the experiments described below the temperature of the developer was maintained at 18° C.

§ 3. THE ABSORPTION OF γ RAYS FROM RADIUM (B+C) IN THIN LAYERS OF ALUMINIUM, COPPER, TIN AND LEAD

The absorption in the above four metals was measured before the rays were finally exponentially absorbed. The source was a single glass tube containing about 100 mg. of radium and was supported in front of a wooden block. The filtering of the rays was equivalent to that effected by 6 mm. of aluminium. With one exception the film used was Agfa. Lead intensifying foils were used throughout.

For each thickness of absorber two films were used, and the time of exposure was chosen to produce a density of approximately 0·6. The time to produce a density of 0·6 could then be calculated, and the reciprocal of this time could be used to give a measure of the intensity of the radiations falling on the film. If the logarithm of the intensity was plotted against the thickness of absorber, the curve became straight when the absorption was exponential. From the earlier part of the curve, together with the exponential curve extrapolated back to zero thickness, the intensity of the softer component could be obtained. This softer component was found to be exponentially absorbed, except that the first point on this curve, that corresponding to zero thickness, was above the exponential curve and thus indicated the presence of a still softer component. Figure 2 shows such a curve for tin.

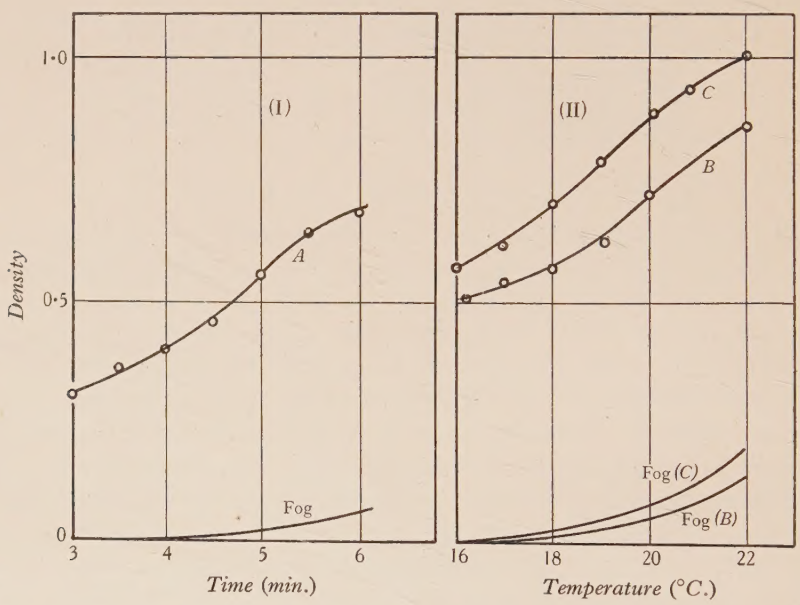

Figure 1. Variation of density with (I) time of development at 18° C.; (II) temperature of developer for time 5 min.

One of the first points that arose from this investigation was that, although the absorption coefficient in aluminium for the harder radiations was the same whether they were filtered in lead or aluminium until the absorption became exponential, the same rule did not hold for copper or tin. Thus the absorption coefficient in copper for γ rays filtered by 1·6 cm. of lead is 0·381 $cm.^{-1}$, but the absorption in copper for rays filtered by copper until the absorption becomes exponential is 0·419 $cm.^{-1}$ For further illustrations of this point see table 5 below.

A summary of the results obtained for the softer radiations is given in table 3. The absorption coefficients μ are given in the second column, the mass absorption coefficients in the third, the ratio of the intensities of the harder to the softer components in the fourth, and the thickness of absorber before the absorption becomes exponential in the fifth. As these absorption coefficients are determined from the

Table 3. Absorption coefficients in aluminium, copper, tin and lead of the softer γ radiations from radium (B + C)

Element	μ ($cm.^{-1}$)	μ/ρ ($cm^2 g^{-1}$)	Intensity ratio harder/softer	d (cm.)	ρd ($g.cm.^{-2}$)
Aluminium	0·48	0·18	2·6	7·4	20
Copper	$2·1_1$	0·24	1·9	2·6	23
Tin	$1·1_7$	0·16	1·5	3·3	24
Lead	$2·5_8$	0·23	1·7	1·6	18

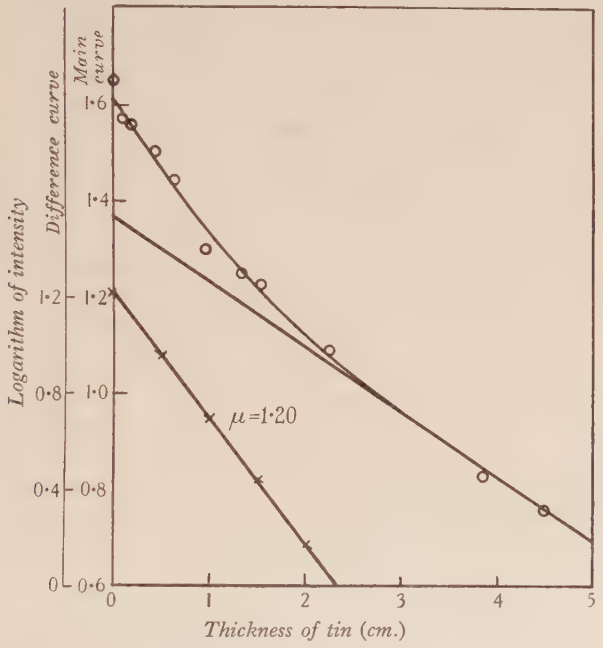

Figure 2. Absorption of γ rays from radium (B + C) in tin.

difference between two curves, the accuracy of determination is not so great as for the harder components. The accuracy of determination in table 3 is about 7 per cent, judging by the departure of individual determinations from the mean. The accuracy of the intensity ratio is limited by the same cause. This ratio is given for zero thickness of absorber, that is to say for a filtering of 6 mm. of aluminium. It will be seen that under these conditions the harder component is approximately twice as intense as the softer. However, if this ratio is to be correct then true intensities, irrespective of wave-length, must be recorded on the film. As mentioned above the photographic action of softer γ rays is intensified approximately equally with that of the harder rays by lead foil when the intensifying action is referred to that of aluminium. This does not, however, establish that the true intensities of all radiations are correctly recorded, for the intensities on the film must be due to the absorption of energy from the beam in the film itself, either directly or through the intensifiers. As the absorption of γ rays decreases with wave-length, the intensity of the shorter wave-lengths will not be fully recorded, and hence the intensity of the harder

component may be greater than twice that of the softer. Kohlrausch[4] has reported a soft component of which the intensity is one-eighth of the hard component. His results were, however, difficult to obtain, as he had a medium component of intensity nearly equal to that of the hard component which arose, as has already been pointed out[1,2], from rays softened in the channel wall used to produce a parallel beam of γ rays.

The values of the absorption coefficients in lead and aluminium can be compared with the results obtained by others. Thus for aluminium Kohlrausch[4] found 0·57 cm.$^{-1}$ for the softer component and Rutherford and Richardson[5] 0·51, compared with the value above of 0·48. The variations are not much greater than the experimental error. For lead the agreement found between the result of Kohlrausch 4·8 and that above 2·6 is not so good.

It is interesting to note the thickness of each absorbing material necessary before the radiations are finally exponentially absorbed. It had been previously pointed out that 1·6 cm. of lead filter produced exponential absorption, whereas others, e.g., Kohlrausch, had found that the thickness of filter required was nearly 4 cm. Clark[6] about the same time as the author had found that the γ rays from radium (B + C) were exponentially absorbed after being filtered by 1·8 cm. of lead. The filter thicknesses d necessary for exponential absorption are given in table 3, and in the last column the product of d and the filter density ρ is seen to be nearly constant. It is, however, difficult to decide the exact point at which the curve becomes exponential, so that there is some uncertainty as to the exact value of ρd. Its approximate constancy indicates that the exponential absorption of the rays is merely a question of the amount of matter to be traversed.

It was pointed out by Kohlrausch and substantiated by the author that μ/ρ for the harder rays, of wave-length 7 X., is approximately constant, μ/ρ being equal to 0·044. Although the experimental values of μ/ρ for the softer component are not nearly so constant, much of the variation may be due to the rather large experimental error in the determination of the absorption coefficients.

Wave-lengths of softer radiations. Some of the effective wave-lengths of the softer radiations can be obtained from an examination of the mass absorption coefficients of X rays of short wave-length. This is done in table 4 below in which the values of μ experimentally determined are given, together with the wave-lengths of the X rays which have the same values of absorption coefficient[7]. The value of the wave-length for tin is somewhat less than 50 X. The effective wave-length for softer radiations in lead is taken from a table prepared by Gentner[8].

Table 4. Effective wave-lengths of softer radiations

Element	Experimental value of μ/ρ	Effective wave-length (X.)	μ/ρ for wave-length
Aluminium	0·18	125	0·178
Copper	0·24	72	0·23
Tin	0·16	<50	0·32
Lead	0·23	31	

It will be seen that the effective wave-length of the softer component decreases with the atomic number of the absorbing material.

§ 4. THE VALUES OF THE ABSORPTION COEFFICIENTS FOR THE HARDER γ RADIATIONS FROM RADIUM (B+C) WITH DIFFERENT FILTERINGS

When it was found for tin or copper that the value of the absorption coefficient depended on whether the γ rays were filtered by lead or by the element in question until the absorption became exponential, a series of experiments was carried out in which the absorption coefficients in aluminium, copper, tin and lead were obtained

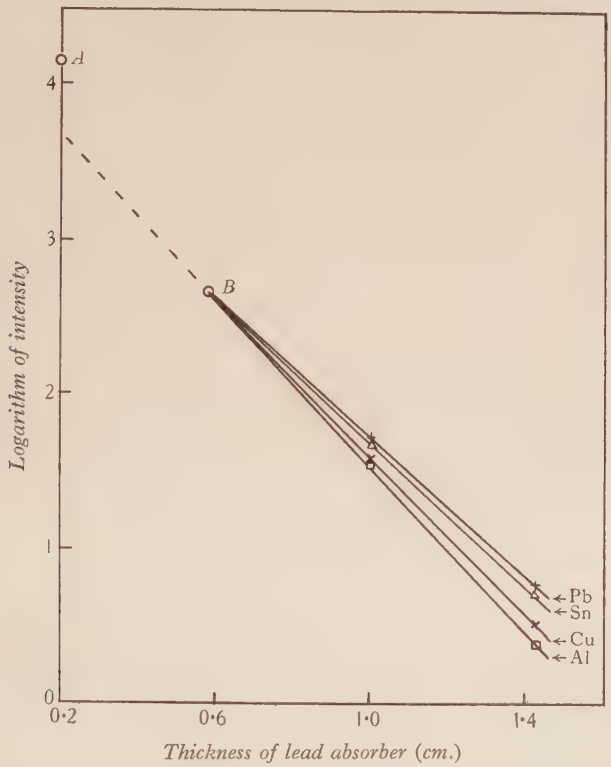

Figure 3. Absorption in lead of γ rays from radium (B+C), previously made exponential in various filters. All curves have been adjusted to pass through point *B*. Point *A* is for aluminium-filtered rays.

with γ rays filtered by aluminium, copper, tin and lead. The source was 400 mg. of radium in four tubes arranged in line. The values of the absorption coefficients are set out in table 5 and the results for lead are shown in figure 3. The thicknesses of the filters are as set out in column 5 of table 3, and the absorbing thicknesses were aluminium 10 cm., copper 3 cm., tin 4 cm., lead 1·4 cm. At least three points were used on each curve to determine the slope.

In these experiments the photographic action was intensified by Patterson

screens, except for the determinations with lead-filtered rays which had been obtained previously with lead-foil intensifying-screens. The accuracy of the determinations is given as the mean departure from the mean result and varies between 2 and 4 per cent.

Table 5. Absorption coefficients with different filterings

Filter \ Absorber	Aluminium	Copper	Tin	Lead
Aluminium	0·124 ± 0·004	0·399 ± 0·010	0·341 ± 0·013	0·606 ± 0·023
Copper	0·123 ± 0·002	0·419 ± 0·016	0·320 ± 0·002	0·581 ± 0·016
Tin	0·119 ± 0·002	0·387 ± 0·018	0·316 ± 0·010	0·554 ± 0·02
Lead	0·123	0·381	0·293	0·533

As can be seen from the table, the absorption coefficients determined in aluminium do not depend on the nature of the filter, but for tin and lead the nature of the filter has a marked influence on the absorption coefficient, in that the lower the atomic number of the filter the greater is the absorption coefficient, i.e., the longer is the effective wave-length. It was found for rays filtered with aluminium, copper and tin that the points for zero thickness of lead used as absorber did not fall on the exponential straight line, and the curve becomes exponential only after a thickness of 5 mm. of lead. (The point for 2 mm. of lead is shown for aluminium-filtered rays in figure 3.) This applies also for the absorption in tin of γ rays filtered with aluminium, while for the remainder the point of zero thickness of absorber falls on the exponential curve.

Somewhat similar filtering effects were found by Ahmad[9] for the γ rays from radium (B + C) in filters of iron, lead and mercury, the thicknesses of the filters being 6·5 cm. of iron, 1 cm. of lead and 3·41 cm. of mercury. The results here obtained for copper and lead can be compared with Ahmad's results for iron and mercury, since the product ρd for these are 51 (iron) and 46 (mercury). As the atomic number of iron is 26 and that of copper 29, the results of iron-filtering should be comparable with those of copper-filtering, as should those of mercury (80) with lead (82). The comparison of Ahmad's results with the author's are set out in table 6.

Table 6. Variation in value of absorption coefficient with filtering

Absorber	$\frac{\mu \text{ with iron filter}}{\mu \text{ with mercury filter (Ahmad)}}$	$\frac{\mu \text{ with copper filter}}{\mu \text{ with lead filter (Rogers)}}$
Aluminium	1·05	1·00
Copper	1·06	1·10
Tin	1·09	1·09
Lead	1·12	1·09

An examination of the results indicates a general agreement in that the filter with the lower atomic number produces a greater absorption coefficient. The numerical agreement is moderately good except that Ahmad finds a variation in aluminium whereas none is found here.

§ 5. DISCUSSION OF RESULTS

In spite of the heterogeneous nature of the γ rays from radium (B + C) the experiments indicate that these rays can be analysed into two apparently homogeneous components in that each appears to be exponentially absorbed. If, however, the mixed beam is passed through increasing thicknesses of an element until it becomes exponentially absorbed in further thicknesses of the element, the effective wave-length of the filtered beam is shorter the higher the atomic number of the filtering element. Thus if aluminium-filtered and lead-filtered rays are separately absorbed in lead, the absorption coefficients are respectively 0·606 and 0·533 cm.$^{-1}$ Further, the effective wave-length of the softer component of the γ rays, as determined from their absorption coefficients, is shorter the higher the atomic number of the absorbing material.

These results can be explained at least approximately by considering the reduction in intensity of the main γ-ray lines in the spectrum of radium (B + C) as the rays pass through matter.* The present discussion will be limited to a consideration of the absorption in aluminium and lead, the elements of least and greatest atomic number that have been investigated.

The absorption of γ rays takes place by three main processes, namely (i) Compton scattering, which is taken into consideration by the Klein-Nishina formula; (ii) the photoelectric effect, which for shorter-wave-length γ rays can be calculated from the results of Hulme and others[10], and (iii) pair-production. The last-named process is inoperative for wave-lengths greater than 12 X. For wave-lengths shorter than this in the known γ-ray spectrum of radium (B + C) it assumes importance only for elements of high atomic number. Thus in the absorption by lead of γ rays of wave-length 7 X., pair-production accounts for 5 per cent only of the total absorption.

The more intense lines in the γ-ray spectrum of radium (B + C) are given in table 7. The intensities of most of the lines are those determined by Ellis and Aston[11]. The intensities for the first two lines of the table are obtained from unpublished work on the γ-ray spectrum. The line 160 X. (really two lines 160 and 165 X.) is photographically the most intense line of the spectrum and has been allotted intensity 1, while the intense lines near 140 X. have been allotted intensity 0·45.

If the absorption coefficients for these different wave-lengths are known, the effective absorption coefficient μ for a thickness d cm. of absorber can be calculated from the equation

$$\mu = \frac{1}{d} \log \frac{\Sigma I'_0}{\Sigma I'_0 e^{-\mu' d}},$$

where the I'_0 terms are the incident intensities and the μ's are the corresponding absorption coefficients.

The absorption coefficients for both aluminium and lead can be obtained for wave-lengths greater than 50 X. from Allen's determination with X rays. For lead

* As the spectrum is heterogeneous it would, however, be anticipated that there would be no exponential absorption for the beam as a whole, for with increasing thickness of absorber the absorption coefficient should steadily decrease.

the absorption coefficients for wave-lengths less than this have recently been summarized by Gentner.[8]* For aluminium the Klein-Nishina formula gives, for wave-lengths less than 50 X. the absorption coefficients with sufficient accuracy for the present purpose; thus for 36·3 X. the photoelectric part of the absorption in aluminium is approximately $\frac{1}{2}$ per cent of that due to Compton scattering. The absorption coefficients used in the calculation are set out in table 7.

Table 7. The more intense lines of the γ-ray spectrum of radium (B + C) and their absorption coefficients in aluminium and lead

Wave-length (X.)	Intensity	μ in aluminium ($cm.^{-1}$)	μ in lead ($cm.^{-1}$)
(160)	(1)	0·577	—
(140)	(0·45)	0·527	—
50·8	0·115	0·311	11·5
41·6	0·258	0·279	4·56
34·9	0·450	0·255	3·16
20·2	0·658	0·213	1·31
10·9	0·206	0·154	0·72
6·94	0·258	0·123	0·53
5·57	0·074	0·107	0·49

It will be seen that, for wave-lengths greater than 50 X., the absorption coefficients are so great, owing to the great importance of the photoelectric absorption, that radiation of these wave-lengths will be reduced to negligible intensity in very small thicknesses of lead.

The calculated effective absorption coefficients for the γ-ray lines given in table 7 are set out in table 8.

Table 8. Effective absorption coefficients ($cm.^{-1}$)

Filter	Absorber	
	1·4 cm. lead	6·0 cm. aluminium
1·6 cm. lead	0·65	0·149
7·4 cm. aluminium	0·91	0·178

It will be seen that in lead there is an excess of 40 per cent in the absorption coefficient for aluminium-filtered rays over lead-filtered rays. In aluminium the two absorption coefficients are more nearly equal, the excess being 19 per cent; experimentally no excess is found. It will be noted, however, that all the absorption coefficients are considerably higher than those found experimentally; see table 5 above. This would be accounted for if there are γ-ray lines of moderate intensity of shorter wave-length than those given in table 7. There is some experimental evidence for the presence of such lines, for Skobeltzyn[12] has found from tracks of recoil electrons that these are γ rays with energies of 3×10^6 electron-volts, whereas the rays of highest energy in the table are $2{\cdot}2 \times 10^6$ eV. Steadman[13] has also found

* There is, however, a discrepancy between the values for lead for shortest wave-lengths found by Allen and the longest wave-lengths summarized by Gentner.

evidence of γ rays of shorter wave-length than those given in the table. Not only will the presence of such short-wave-length lines reduce the values of the absorption coefficients, but also it will make the absorption coefficients in aluminium of differently filtered rays more nearly equal. Thus if the presence of a line of 4·65 X.* of intensity 0·4 in the scale of intensities of table 7 is assumed, the values of the absorption coefficients of lead-filtered rays are 0·58 $cm.^{-1}$ in lead and 0·126 in aluminium, and the coefficients for aluminium-filtered rays are 0·72 in lead and 0·147 in aluminium. For all the values here are much nearer the experimental values than those in table 8 so that there must be γ-ray lines of considerable intensity having wave-lengths shorter than 5·6 X.

Finally from the calculated values in table 8 for the absorption coefficients of the more penetrating radiations those of the softer radiations also can be calculated. It is necessary, in carrying out this calculation for aluminium, to take into consideration the lines of 160 and 140 X. The calculated absorption coefficient is then 0·57 $cm.^{-1}$, which is somewhat higher than the experimental value of 0·48. The lines of 160 and 140 X. need not be considered for lead, for they are reduced to negligible intensity in considerably less than 1 mm. of lead; moreover in determining the experimental values of the absorption coefficient the point for zero thickness of lead was not included. The calculated absorption coefficient for the softer radiations in lead is found to be 2·6 $cm.^{-1}$, which is very close to the experimental value.

§6. ACKNOWLEDGMENTS

These experiments were carried out in the Commonwealth Radium Laboratory, University of Melbourne, and the author wishes to express his thanks to Mr A. H. Turner, M.Sc. of that laboratory for making available its facilities. He also is grateful to Dr L. H. Martin, with whom certain points in this paper have been discussed.

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* This particular wave-length was assumed to illustrate the point as its absorption coefficients are accurately known.

THE EFFECT OF HYDROGEN ON THE TIME-LAG OF ARGON-FILLED PHOTOELECTRIC CELLS

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ABSTRACT. In a previous paper of this series[1] it was stated that the addition of small quantities of hydrogen to the argon filling of a photoelectric cell with unsensitized potassium cathode reduced the time-lag greatly. The amount of hydrogen necessary for this effect has now been determined roughly. A Pirani gauge has been found very suitable for determining a small proportion of hydrogen in argon.

§ 1. APPARATUS

The previous paper of this series[1] left undetermined the proportion of hydrogen required to produce the decrease in time-lag described in § 5; it was thought that somewhat elaborate apparatus would be required to determine the proportion of hydrogen present in argon at low pressure.

It has been found that the problem can be solved very simply and with sufficient accuracy by the use of a Pirani gauge. The gauge was of the simple type in which a constant p.d. is maintained across the bridge, and the out-of-balance current is read. Figure 1 shows typical calibration curves obtained against a McLeod gauge. The thick line refers to pure argon; the thin lines terminating on it refer to mixtures of argon and hydrogen in which the argon pressure is constant. It will be seen that the various thin lines are all very similar, so that it is easy to interpolate between them.

Accordingly all that is necessary is to attach such a gauge to the cell, fill it with argon to a suitable pressure, and draw the thin line in figure 1 terminating on the thick line at this pressure. When the reading of the gauge is varied by adding hydrogen (through a palladium tube) or withdrawing it (through a palladium tube or by absorption at the potassium surface), the corresponding pressure, and therefore the proportion of hydrogen, is read off on this thin line. Of course it is assumed that there is no absorption, or other change, affecting the quantity of free argon; everything pointed to the truth of this assumption.

§ 2. RESULTS

The results obtained by this method can be stated thus, the pressure of argon in the cell being about 0·2 mm. The effect of introducing a small proportion of hydrogen into argon is (1) to decrease the magnification at a given voltage, (2) to decrease the time-lag at a given magnification and therefore, a fortiori, at a given voltage. Both

these effects increase rapidly as the amount of hydrogen is increased up to (say) 2 per cent by volume; the rate of increase then falls off and becomes negligible at about 6 per cent. Accordingly the statement made previously that the effect is produced by *small* proportions of hydrogen is correct. The exact variation of the effects with amount of hydrogen is complicated and depends on the nature of the cell; it is useless to describe the experiments in detail until some coordinating theory can be suggested.

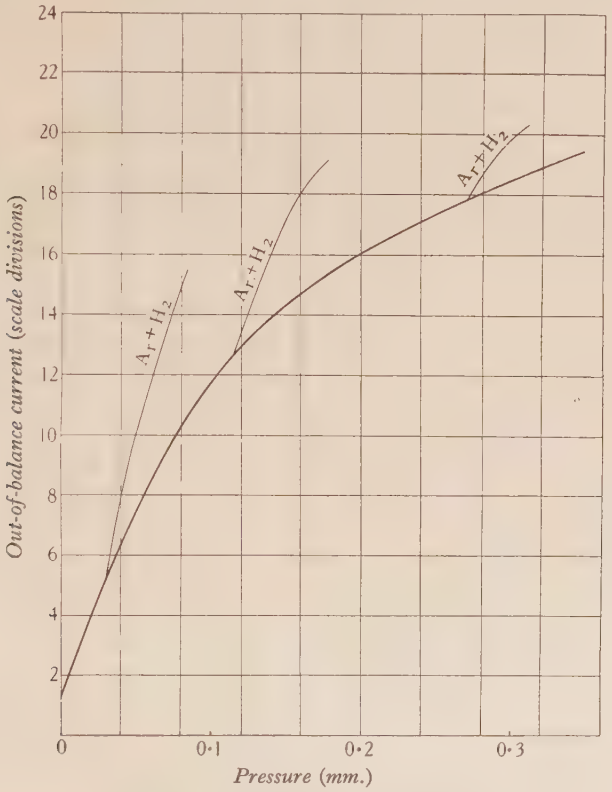

Figure 1. Calibration of Pirani gauge.

Effect (1) is already known[2]. Effect (2) is that described in the previous paper. A third effect described in that paper, namely a decrease in the emission from the potassium, was not obtained again; it doubtless depends on the extent to which the potassium has already adsorbed or absorbed hydrogen.

It is to be observed that effects (1) and (2) were correlated with hydrogen free in the cell. It is conceivable that the direct cause was hydrogen absorbed on the cathode in an amount which is in equilibrium with the amount free in the cell. But nothing was observed that confirms that suggestion.

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THERMOMAGNETIC PROPERTIES OF NICKEL WIRE, PART 3

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ABSTRACT. The paper constitutes a brief report of results of a complete and careful analysis of observations previously published on the e.m.fs. found in nickel wires.

RESULTS published previously in parts 1 and 2[1] have been subjected to a careful and detailed analysis of all corrections, the homogeneous e.m.fs. measured in part 2 being employed in the corrections to the results in part 1. The results of this analysis, details of which have to be omitted owing to lack of space, are best exhibited graphically. The figures given are self-explanatory.

The most important points brought out by these curves are: (*a*) There is a constant potential gradient susceptibility remaining in the material at temperatures well above the Curie point; (*b*) there is a Villari reversal phenomenon of the potential-gradient susceptibility under different tensional strains; (*c*) the magnetic change of the Thomson specific heat is found to vanish, under all tensions, at a definite Curie point which coincides with the temperature at which the potential-gradient susceptibility becomes independent of temperature.

The first two points are interpreted as showing that the magnetic change in potential-gradient is not entirely ferromagnetic in origin: the ferromagnetism of nickel is known not to show any Villari reversal, and to vanish at the Curie point. They can also be interpreted in terms of Bridgman's theory of thermoelectricity.

According to Bridgman's theory the e.m.f. E_{ab} in the circuit is not, as is usually assumed, due to a combination of contact and internal e.m.fs., the Peltier heat being in fact accompanied by no driving e.m.f. The total driving e.m.f. in the circuit has to be derived from the formula[2]

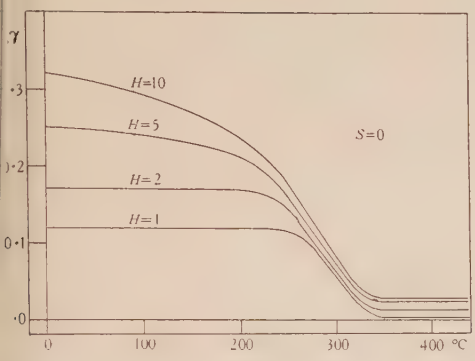
$$\delta E = -\delta T \left[\int_0^T (\sigma/T)\, dT + f(T) \right] \qquad \ldots\ldots(1),$$

where $f(T)$ is some function identical for all metals and contributing nothing to the total e.m.f. in any circuit, which works out as

$$E_{ab} = \int_{T_1}^{T_2} \Delta\gamma . dT \qquad \ldots\ldots(2),$$

where

$$\Delta\gamma = -\int_0^T \{(\sigma_a - \sigma_b)/T\} . dT \qquad \ldots\ldots(3).$$



ure 1. Thomson potential-gradient susceptibility. Changes in Thomson potential-gradient due to fields under 1, 2, 5, 10 amperes current at zero tension as functions of temperature. (10 amperes gave 303 G.)

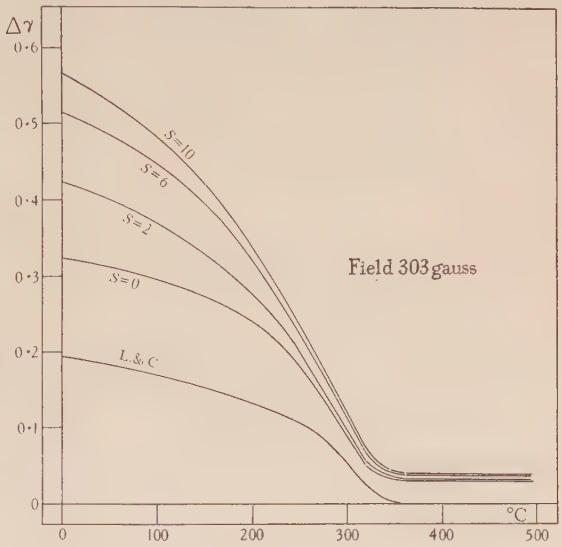
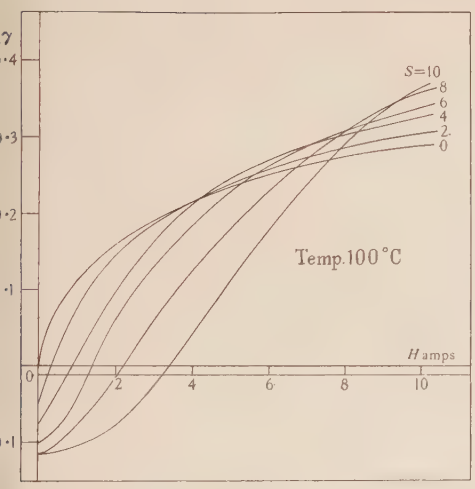


Figure 2. Thomson potential-gradient susceptibility. Changes in Thomson potential-gradient due to a field of 303 G., under tensions of 0, 2, 6, 10 kg., as function of temperature. (1 kg. gave 127·3 kg./cm².)



ure 3. Relationship between Thomson potential-gradient, magnetization and strain. At a mean temperature of 100° C. the change produced in the Thomson potential-gradient as a function of magnetic field for tensions of 0, 2, 4, 6, 8, 10 kg.

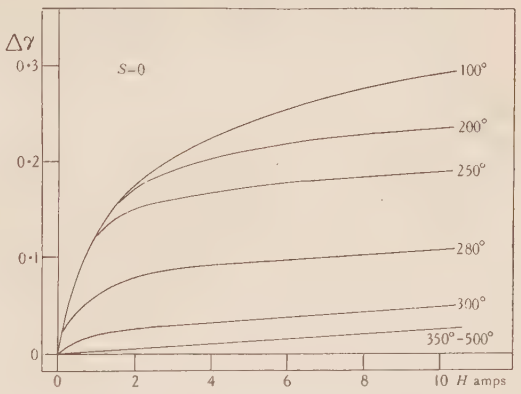


Figure 4. Relationship between Thomson potential-gradient, magnetization and temperature. At zero tension the change in Thomson potential-gradient as function of magnetic field for temperatures of 100, 200, 250, 280, 300 and between 350–500° C.

The coefficient γ is evidently what is generally known as the Thomson potential-gradient.

The constant values of $\Delta\gamma$ found at temperatures above the Curie point are the

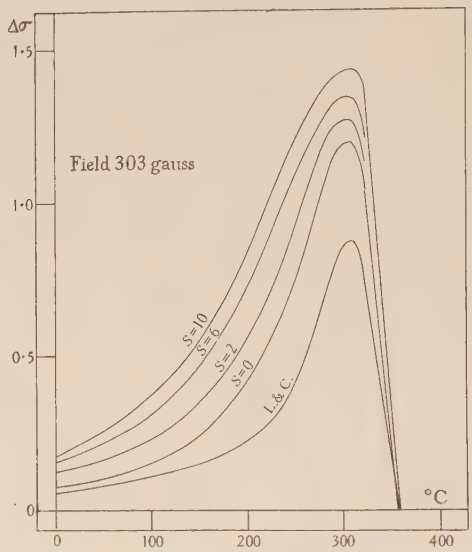


Figure 5. Thomson heat susceptibility. The change in Thomson specific heat, calculated by the Kelvin formula from the gradients of our curves, and due to a field of 303 G. under tensions 0, 2, 6, 10 kg. as a function of temperature. The curve marked "L. & C." has been drawn from data published by Lowance and Constant[3].

values of the integral in equation (3) when T is put equal to the Curie-point temperature. There is no mathematical reason why these should be zero.

No explanation has been found for the antisymmetrical part of the e.m.fs. examined in part II.

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X-RAY INVESTIGATION OF PURE IRON-NICKEL ALLOYS. PART 1: THERMAL EXPANSION OF ALLOYS RICH IN NICKEL

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ABSTRACT. The thermal expansion of pure iron-nickel alloys having compositions ranging between 97 and 73 atoms of nickel per cent was measured by the X-ray method. Close agreement was found between the expansion of the crystal lattices and the expansion of specimens of material consisting of composite masses of crystals, such as were used by previous workers who employed the ordinary methods of measurement. For the alloys examined, which were all rich in nickel, the addition of iron to the nickel lattice was found to have two effects: (*a*) the nickel lattice expanded approximately in proportion to the amount of iron added, and (*b*) the temperature of the discontinuity in the thermal-expansion curve was raised with the addition of iron, but not in proportion to the amount of iron added. No change in structure took place at the temperature of magnetic transformation; the face-centred structure persisted from room-temperature through the transformation temperature up to 600° C., the highest temperature reached in the investigation. No ageing effect was observed with an alloy containing 94 atoms of nickel per cent, the lattice parameter showing no consistent change over a period of 15 months.

§ 1. INTRODUCTION

Owing to their importance in industry, the iron-nickel alloys have been extensively investigated. They have been found to possess many interesting and unusual properties. The presence of small quantities of impurities considerably alters their properties and gives rise to phenomena which are difficult to follow, especially as the quantities of impurities involved may be very small. To avoid complications of this nature it was decided in the first instance to investigate alloys made from the pure elements in the hope that the results thus obtained would form the basis for further work on alloys containing small amounts of a third constituent.

Previous investigators have found that all the alloys solidify as solid solutions which upon cooling suffer both phase and magnetic transformations. It is upon these transformations that many of the peculiar properties of the alloys depend.

In the present investigation, an account of which will be given in this and subsequent papers, a detailed examination was made of the lattice dimensions and structures at different temperatures with the object of throwing more light if

possible on the transformations that occur and of studying the thermal expansion of the alloys in its relation to these transformations.

This paper deals with the thermal expansion of five alloys rich in nickel.

§ 2. HEAT TREATMENT AND PREPARATION OF THE ALLOYS

The alloys were kindly provided by the Mond Nickel Company, Ltd.; their compositions are given in table 1.

Table 1. Composition of alloys

Alloy	Nickel-content	
	Weight per cent	Atoms per cent
97	96·75	96·6
94	93·95	93·7
91	91·04	90·7
83	82·86	82·2
74	74·00	73·0

Although the alloys had been lump-annealed at 1150° C. for 20 hours in a nitrogen atmosphere before they were received, they were submitted to further heat treatment before powders were prepared from them. For this purpose a small lump of each alloy was enclosed in a pyrex glass tube which was evacuated and sealed. Each tube, wrapped in asbestos cloth, was placed in a furnace maintained at 800° C. and was allowed to remain at that temperature for a number of days before being cooled slowly to the temperature of the room. Measurements made on alloy 74 showed that this further heat treatment was not necessary because no appreciable difference in the lattice parameter was observed between specimens which had received no lump annealing and others which had received respectively 3, 6 or 12 days' annealing at 800° C. The initial heating which the ingots had received was therefore adequate to produce uniformity of composition.

Filings taken from the ingots were annealed to remove distortion due to filing. Care was taken in the filing operation not to apply too great a pressure, in order to minimize contamination with particles torn from the file. The filings were passed through a 100-mesh sieve before being sealed in evacuated pyrex glass tubes. These tubes after sealing were placed in deep holes in an iron block which fitted into the furnace; the temperature was recorded by a calibrated thermocouple placed in close contact with the middle of the block. The annealing was then carried out according to plan.

Difficulty was experienced with some of the alloys in obtaining spectral lines suitable for accurate measurement. When filings of alloy 74, for example, were annealed at 300° C. for 10½ hours and cooled moderately fast in air, the lattice was still too distorted to yield spectral lines. The distortion might be due to the cold work done in filing, or to the rapid cooling to room-temperature. On the assumption that both effects were present, the filings were annealed at a higher temperature of 800° C. for 2 hours and were allowed 90 hours to cool to room temperature. After this treatment good lines were obtained, but they showed that the material was now

over-annealed. In an attempt to find the conditions necessary to produce the best reflection lines, some more powder was annealed at 500° C. for 12 hours and cooled over a further period of 33 hours. A marked improvement in the definition of the lines was observed. These observations showed that the annealing at 800° C. was too severe, while that at 300° C. was insufficient to remove the effects of cold work.

It was found during the course of the work on alloy 74 that not only the temperature of annealing is important, but also the rate of cooling from this temperature to room-temperature. A powder heated at 600° C. may yield good lines at that temperature, but no lines at room-temperature if it is cooled in too short a time. The necessity for a slow rate of cooling after annealing becomes less as the nickel-content of the alloy increases, as good lines being obtained for quenched as for slowly cooled specimens of pure nickel.

The annealing treatment given to the alloys described in this paper is recorded in detail in table 2.

Table 2

Alloy no.	Lump annealing			Powder annealing		
	Temperature (°C.)	Time (days)	Period of cooling (days)	Temperature (°C.)	Time (hours)	Period of cooling (hours)
97 (1)	800	6	—	690	12	27
(2)	800	6	—	500	14	24
94 (1)	800	6	—	690	12	27
(2)	800	6	—	500	14	24
(3)	800	6	—	600	15	225
91 (1)	800	6	—	690	12	27
(2)	800	6	—	500	14	24
(3)	800	6	—	600	15	144
(4)	800	6	—	600	7	108
(5)	800	6	—	700	12	29
83 (1)	800	6	—	690	12	27
(2)	800	6	—	500	14	24
74 (0)	—	—	—	300	$10\frac{1}{2}$	—
(1)	—	—	—	800	2	90
(2*a*)	860	3	—	800	2	90
(2*b*)	860	3	—	500	12	33
(3*a*)	860	$6\frac{1}{4}$	—	800	2	90
(3*b*)	860	$6\frac{1}{4}$	—	500	12	33
(4)	800	12	$4\frac{1}{2}$	600	14	48

§ 3. PROCEDURE

The high-temperature precision camera employed in this work was made to fit inside a strong airtight box and all exposures at temperatures above that of the room were made *in vacuo*. The vacuum was maintained by keeping a rotary oil pump connected to the sealed chamber, an oil-trap immersed in freezing mixture being inserted between the two.

The powder to be studied was cemented to a specially prepared copper sheet about 2 mm. thick, to the face of which one junction of a calibrated thermocouple was attached with silver solder. This sheet was held firmly in position on the camera by means of thin copper foil and springs; it was separated from the camera by a

thin sheet of mica. Before proceeding it was important to investigate whether the adhesive used to mount the specimen on the thermo-sheet had any effect on the lattice parameter of the alloy. The adhesive, which was a dilute solution of seccotine, charred at about 300° C. and there was risk that the residue might combine with the alloy at the higher temperatures reached.

An attempt was made to eliminate the adhesive by pressing the powder into V-shaped grooves cut in the face of the thermo-sheet in such a way that the powder could be made to lie evenly in them when the thermo-sheet was placed vertically in the chamber. With this arrangement it was found that the temperature of the powder was lower than that recorded by the thermocouple.

The next attempt to eliminate the adhesive necessitated the rearrangement of the whole apparatus so that the X-rays fell vertically from above on the thermo-sheet, which was now horizontal, with its concave surface upwards. In this position, the powder could be sprinkled evenly on the sheet without any adhesive having to be used. The temperature of the powder was again lower than that indicated by the thermocouple. Exposures made with silver powder showed that the temperatures were in error by as much as 10 per cent. To take one example, when the silver powder was at a temperature of 321° C., deduced from the value of its parameter, the thermo-sheet upon which it lay was at a temperature of 350° C. On cementing the powder to the thermo-sheet, the discrepancy disappeared. The results of some of the experiments made in this direction are recorded in table 3.

Table 3

Film no.	Temperature as given by thermo-sheet (°C.)	Lattice parameter of silver (A.)	Temperature as given by the lattice parameter	
B 3	350	$4{\cdot}1023_2$	321	No adhesive
B 4	353	$4{\cdot}1015_5$	313	,,
B 5	351	$4{\cdot}1046_2$	348	With adhesive
B 6	350	$4{\cdot}1036_2$	336	No adhesive
B 7	352	$4{\cdot}1054_7$	356	With adhesive
B 8	506	$4{\cdot}1192_2$	508	,,

The temperatures given in the fourth column were obtained from the {lattice-parameter, temperature} curve of silver, which had been determined previously. It is evident from these results that the adhesive serves the double purpose of holding the powder in position and of ensuring good thermal contact between the powder and the thermo-sheet.

It was found, when a specimen was used to obtain a series of photographs at high temperatures, and exposures at room temperature were interspersed among these, that the parameter at room temperature remained constant if the powder retained its metallic lustre. This was evidence that the charred adhesive did not combine with the alloy to any measurable extent at the temperatures reached.

It was noticed in the preliminary stages of the work that some of the alloys lost their metallic lustre on being heated in the evacuated chamber, but the adhesive was

not responsible for this. A thorough investigation into the cause of the tarnish was essential before proceeding further. As a first step a polished sheet of copper was left for three days in the evacuated chamber, and at the end of this period the sheet was found to be tarnished although it had remained at room-temperature all the time. The tarnish was of a sulphurous nature, and steps were therefore taken to rid all parts of the apparatus of sulphur and water vapour. The walls of the enclosure were carefully cleaned with ammonia, and the rubber washers and pressure-tubing connexions were boiled in caustic soda. Finally two small dishes, one containing potassium hydroxide and the other phosphorous pentoxide, were inserted in the enclosure, which as has already been stated was separated from the oil-pump by an oil-trap immersed in ice. With these precautions the discoloration of the powder did not occur, but it was found necessary to maintain a pressure in the chamber not exceeding 0·001 mm. of mercury.

All the photographs taken after this were obtained with powder held in position on the thermo-sheet by a thin layer of adhesive, and the above precautions were taken to remove from the chamber any gases and vapours that might be likely to affect the specimen.

The length of time during which the powder when mounted on the camera was kept at the temperature of exposure before the actual exposure was made varied between wide limits. It was found with the alloys now under investigation that heating for half an hour before the exposure gave the same result as heating for much longer periods. The powders before being exposed at high temperature to X-rays were subjected to three different heat treatments. (1) The powder was maintained at the required temperature for half an hour or more; unless the contrary is stated, this method was the one usually adopted. (2) The powder was heated rapidly to a temperature 100° C. lower than that at which the exposure was to be made and gradually brought up to the temperature of exposure over a period of at least half an hour; (*h*) in the tables that follow indicates the use of this method. (3) The material was heated to a temperature 100° C. higher than that at which the exposure was required and was allowed to cool slowly to the temperature of exposure over a period of at least half an hour; (*c*) indicates the use of this method. The temperature during the exposure was always kept steady.

The satisfactory agreement between the values of the lattice parameter of a given specimen obtained by these three methods shows that the temperature could be accurately controlled and determined. The high-temperature exposures were not made in order of increasing or decreasing temperature; a random sequence was adopted in order to eliminate progressive temperature errors. It was always arranged, when high-temperature photographs were taken with a given specimen, that a few photographs at ordinary temperature should be taken to ascertain whether the lattice returned to its normal dimensions. This precaution served as a useful check on the state of the material after heating; if by any chance the vacuum chamber became contaminated with vapours which at the higher temperatures reacted vigorously with the alloy, this reading taken at room temperature would at once demonstrate the fact and the material would have to be discarded.

§ 4. SOURCES OF ERROR

There is no need to discuss in detail the sources of error in the determination of lattice parameters with the precision camera, as these have been considered in previous papers. It may however be pointed out that when the same camera, radiation, and film-measuring instrument are used throughout a series of lattice-parameter determinations of the same alloy, and when precautions are taken to keep the conditions of exposure constant, an accuracy of about 0·004 per cent is attainable in an individual result relative to others in the same series of lattice-parameter determinations. An accuracy greater than 0·007 per cent cannot be claimed for the absolute value of any single lattice-parameter determination.

By frequently checking the thermocouple calibration, and by measuring the lattice parameters of silver at definite temperatures, the error in the temperature-determination was reduced to less than 1 per cent. The thermal expansion of silver had been the subject of previous investigation and it was then pointed out that the lattice-parameter values of this metal can be conveniently employed as a check on the temperature-determinations.

Owing to the limits set by the accuracy of the lattice-parameter measurements, the coefficient of thermal expansion cannot be determined to an accuracy greater than about 4 per cent. If several determinations are made, the accuracy of the mean will of course be higher than this. In addition the method yields a {lattice-parameter, temperature} curve which serves a useful purpose in that by its means any irregularities are immediately detected.

One point in connexion with the determination of coefficients of thermal expansion of materials by the X-ray method should be emphasized. With the precision camera employed it would be impossible to obtain well-defined lines and resolved doublets if any distortion existed in the material examined. In consequence the specimen has to be carefully heat-treated until it yields well-defined lines even at the largest glancing angles (about 85°) possible with the camera. In these circumstances the values of the thermal coefficients apply to material which is entirely free from internal stresses.

The K characteristic radiations of copper and manganese were employed. Copper radiation was reflected in the (331) and (420) planes and was used with alloys 74 and 83; manganese radiation was reflected in the (311) planes and was used with alloys 91, 94 and 97. This latter radiation was obtained by the electro-deposition of manganese on a copper target by immersion in a weakly acid solution of manganous chloride. The wave-lengths employed in the calculations were: $CuK\alpha_1$, 1·537395 A.; $CuK\alpha_2$, 1·541232 A.; $MnK\alpha_1$, 2·09751 A. and $MnK\alpha_2$, 2·10149 A.

§ 5. RESULTS

Alloys 97, 94 *and* 91. The use of manganese radiation in the investigation of the thermal expansion of these three alloys made it possible to obtain lines in favourable positions on the films for accurate determination of the parameters. The results obtained are collected in table 4.

Table 4

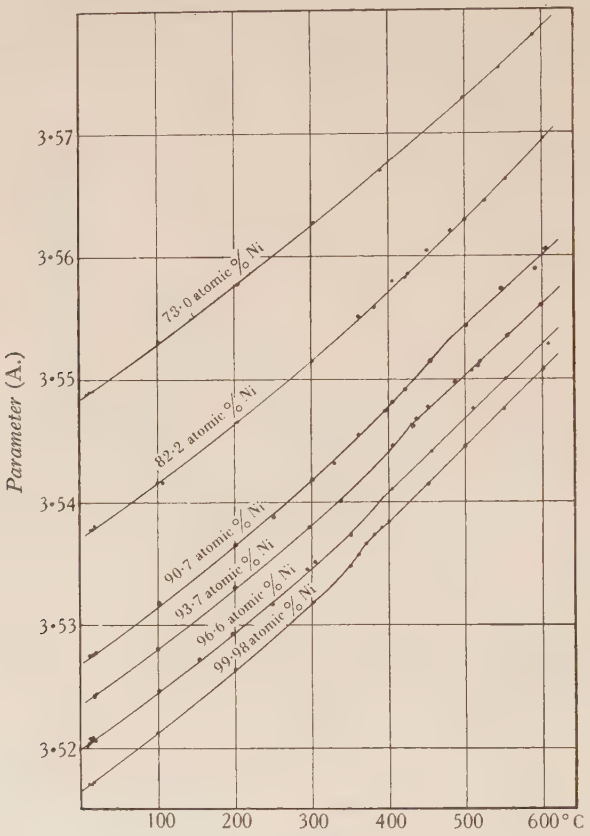
Alloy ... 97			94			91		
Nickel-content (per cent by weight) 96·75			93·95			91·04		
Film no.	Temperature (°C.)	Lattice parameter (A.)	Film no.	Temperature (°C.)	Lattice parameter (A.)	Film no.	Temperature (°C.)	Lattice parameter (A.)
E 536	11	$3{\cdot}5204_1$	E 515	16	$3{\cdot}5242_1$	V 1	12	$3{\cdot}5275_7$
E 531	13	$3{\cdot}5208_2$	V 37, E 510	17	$3{\cdot}5241_8$	E 489, E 494	16	$3{\cdot}5277_8$
E 537, E 538	14	$3{\cdot}5206_2$	V 34	18	$3{\cdot}5243_4$	E 493	17	$3{\cdot}5277_9$
E 523	16	$3{\cdot}5207_9$	E 495	19	$3{\cdot}5240_7$	V 22, V 27	21	$3{\cdot}5279_4$
E 519	17	$3{\cdot}5209_2$	E 496, E 504	20	$3{\cdot}5243_9$	V 24	22	$3{\cdot}5281_3$
E 530	20	$3{\cdot}5206_4$	E 513	101 (*h*)	$3{\cdot}5280_6$	V 20	102 (*c*)	$3{\cdot}5318_1$
E 524	101 (*h*)	$3{\cdot}5246_7$	E 514	200 (*c*)	$3{\cdot}5331_2$	V 18	201 (*c*)	$3{\cdot}5366_6$
E 532	154 (*h*)	$3{\cdot}5274_6$	V 35	297 (*h*)	$3{\cdot}5379_6$	V 29	249 (*c*)	$3{\cdot}5388_5$
E 525	198 (*c*)	$3{\cdot}5293_5$	E 512	338 (*c*)	$3{\cdot}5400_4$	V 21	301 (*h*)	$3{\cdot}5419_1$
E 529	248 (*c*)	$3{\cdot}5317_0$	E 501	363 (*c*)	$3{\cdot}5420_0$	V 30	328 (*h*)	$3{\cdot}5423_4$
E 534	294 (*h*)	$3{\cdot}5346_1$	E 497	404 (*h*)	$3{\cdot}5446_5$	V 17	360	$3{\cdot}5454_9$
E 520	304 (*h*)	$3{\cdot}5351_5$	E 518	432 (*h*)	$3{\cdot}5462_2$	V 5	396 (*h*)	$3{\cdot}5474_1$
E 527	351 (*c*)	$3{\cdot}5376_5$	E 511	437 (*h*)	$3{\cdot}5467_7$	V 19	403 (*h*)	$3{\cdot}5481_6$
E 522	404 (*h*)	$3{\cdot}5411_6$	E 499	451 (*c*)	$3{\cdot}5477_5$	V 31	422 (*c*)	$3{\cdot}5491_7$
E 533	455 (*h*)	$3{\cdot}5441_4$	E 502	486 (*c*)	$3{\cdot}5498_2$	V 25	454 (*h*)	$3{\cdot}5515_2$
E 521	509 (*c*)	$3{\cdot}5476_2$	V 36	508 (*h*)	$3{\cdot}5507_3$	V 28	501 (*h*)	$3{\cdot}5544_5$
E 535	552 (*c*)	$3{\cdot}5500_0$	E 517	516 (*c*)	$3{\cdot}5510_3$	V 26	546 (*c*)	$3{\cdot}5573_4$
E 526	606 (*h*)	$3{\cdot}5529_3$	E 516	519 (*c*)	$3{\cdot}5514_0$	V 6	591 (*h*)	$3{\cdot}5589_4$
			E 500	554 (*h*)	$3{\cdot}5535_4$	V 23	605	$3{\cdot}5605_1$
			E 503	598 (*h*)	$3{\cdot}5559_7$			

The film numbers included in this table give an indication of the order in which the exposures were made. It is impossible without the aid of a graph to detect the changes that take place at certain temperatures in the three alloys. The figure includes the curves for these three alloys together with the curve for pure nickel[1], which is included for comparison with the other data. In each case a change in slope is noticed, but it occurs at a different temperature with each alloy. For pure nickel it occurs at about 370° C., but it increases to 390° C. for alloy 97, 430° C. for alloy 94 and 490° C. for alloy 91. The discontinuities are accentuated if the slope is plotted against the temperature.

There is a similarity between the thermal expansion of these three iron-nickel alloys and that of pure nickel. The addition of iron to nickel causes the nickel lattice to expand approximately in proportion to the amount of iron added, and it also raises the temperature of the discontinuity observed in the {parameter, temperature} curve. The discontinuity occurs with nickel at the temperature of magnetic transformation. The discontinuities in the other alloys are also closely associated with the magnetic transformation. For instance, Hanson and Hanson[2] found that the thermal arrest temperatures of a 90·71-per-cent alloy were 487° C. during heating and 457° C. during cooling. The discontinuity now found with alloy 91 which

contains 91·04-per-cent nickel occurs at about 490° C. The same sort of agreement is obtained with the other alloys.

The coefficient of thermal expansion of each alloy increases as the temperature is raised, and reaches a maximum value at the temperature of discontinuity, i.e. at the transformation temperature. Approximate values of the coefficients of expansion at various temperatures for the three alloys 97, 94 and 91 are collected in table 5.



Ageing test on alloy 94. In order to make sure that the lattice parameters of the alloys at this end of the equilibrium diagram remained constant over long periods of time, an ageing test was made on alloy 94. The investigation was carried out over a period extending from January 1934 to March 1935. The results of the measurements are summarized in table 6.

These results show that there is no consistent change in the lattice parameter over a total period of 15 months and that in the case of this particular alloy there is no ageing effect.

Alloys 83 *and* 74. Difficulty was experienced with the alloys in obtaining X-ray diffraction lines but this was overcome by cooling the annealed filings slowly instead of rapidly. Even after the right heat treatment for the alloys had been found

the lines obtained were always faint. This effect was noticed also with alloys of higher iron-content.

Table 5

Temperature (°C.)	True coefficient of thermal expansion × 10^6		
	Alloy 97	Alloy 94	Alloy 91
0	12·8	12·8	12·7
100	13·2	13·3	13·4
200	14·0	14·2	14·3
300	16·1	15·9	15·7
380	19·2	—	—
390	19·6	—	—
400	16·7	18·8	17·7
420	—	19·4	—
430	—	19·7	—
440	—	16·5	—
480	—	—	19·7
490	—	—	20·0
500	16·7	16·5	16·4
600	16·7	16·5	16·4

Table 6. Ageing test on alloy 94

Film no.	Specimen	Temperature (°C.)	Lattice parameter (A.)	Date of measurement
E 313	1	14	$3{\cdot}5241_9$	January 5 1934
E 314	2	14	$3{\cdot}5242_2$	
V 34	1	18	$3{\cdot}5243_4$	23 June 1934
E 496	2	20	$3{\cdot}5245_1$	24 July 1934
E 510	3	17	$3{\cdot}5242_8$	9 October 1934
E 509	2	15	$3{\cdot}5243_3$	29 March 1935
E 518	3	14	$3{\cdot}5240_0$	

The investigation of these two alloys was carried out with copper radiation which yielded two doublets; the presence of the two doublets compensated in some measure for the adverse positions of the lines and their poor intensity. In the early stages of the work it was observed that the lattice did not always return to its normal dimensions at room-temperature after being heated to high temperatures. This effect, as in the case of the other alloys, was directly connected with the discoloration of the powder which sometimes occurred at the higher temperatures. Even when every precaution was taken to remove the cause of this discoloration the powders still showed on occasions a tendency to lose their metallic lustre. To minimize the effect the powders were used only once for a high-temperature exposure. In addition a careful examination of the heated powder was made after each exposure and if it showed signs of deterioration the film was discarded.

With both the alloys difficulty was found in obtaining lines between 550 and 600° C. The magnetic transformations for these alloys occur between these two temperatures[2]. The results obtained with the alloys are collected in table 7.

With the exception of the points 606° C. for alloy 74 and those at 553 and 601° C. for alloy 83, the points connecting lattice parameter with temperature lie

Table 7

Alloy ...	83		74		
Nickel-content (per cent by weight)	82·86		74·00		
Film no.	Temperature (°C.)	Lattice parameter (A.)	Film no.	Temperature (°C.)	Lattice parameter (A.)
{E 354, E 369	13	$3{\cdot}5378_5$	E 292	8	$3{\cdot}5488_2$
E 367	14	$3{\cdot}5377_9$	{E 284, E 285, E 286, E 287, E 301, E 302	12	$3{\cdot}5490_0$
E 358	15	$3{\cdot}5377_1$			
E 364	18	$3{\cdot}5381_9$	E 312	15	$3{\cdot}5493_0$
E 365	100	$3{\cdot}5416_7$	E 282, E 296	16	$3{\cdot}5491_3$
E 361	107 (*c*)	$3{\cdot}5415_7$	E 294	101	$3{\cdot}5531_6$
E 356	203 (*h*)	$3{\cdot}5466_2$	E 303	205	$3{\cdot}5577_8$
E 359	302	$3{\cdot}5515_8$	E 293	303	$3{\cdot}5627_7$
E 363	361	$3{\cdot}5551_3$	E 297	390	$3{\cdot}5670_8$
E 379	382 (*h*)	$3{\cdot}5557_9$	E 295	498	$3{\cdot}5728_9$
E 374	383 (*h*)	$3{\cdot}5557_9$	E 306	543	$3{\cdot}5753_1$
E 355	404 (*h*)	$3{\cdot}5580_1$	E 310	587	$3{\cdot}5780_0$
E 376	422 (*h*)	$3{\cdot}5583_0$	E 283	606	$3{\cdot}5803_2$
E 381	425 (*h*)	$3{\cdot}5586_4$			
E 366	450 (*h*)	$3{\cdot}5605_2$			
E 384	481 (*h*)	$3{\cdot}5621_1$			
E 360	500	$3{\cdot}5629_2$			
E 372	524	$3{\cdot}5645_1$			
E 368	553 (*h*)	$3{\cdot}5665_9$			
E 357	601 (*h*)	$3{\cdot}5695_8$			

satisfactorily on smooth curves. The deviations from these curves at the highest temperatures are due probably to the proximity of the magnetic transformation of these alloys. The difficulties encountered at these elevated temperatures restricted the number of observations that could be taken without unduly prolonging the investigation. It is to be noted also that about 600° C. is the working limit of temperature for the camera.

The curves showing the relation between parameter and temperature are included in the figure. The values of the coefficient of thermal expansion at different temperatures calculated from these curves are recorded in table 8.

Table 8

Temperature (°C.)	Coefficient of thermal expansion × 10^6	
	Alloy 83	Alloy 74
0	12·4	12·2
100	13·3	12·9
200	14·2	13·5
300	15·1	14·2
400	16·0	14·9
500	16·9	15·6
600	17·8	16·4

The variation of the true coefficient of thermal expansion with temperature is linear within experimental error between room-temperature and 500° C.

§ 6. CONCLUSIONS

There is a close agreement between the results of the X-ray crystal method of determining the coefficients of thermal expansion and those obtained with methods employed by other workers, notably Guillaume[3] and Chevenard[4], who used larger masses of material. This fact demonstrates again that the metal as a whole, consisting of a composite mass of crystals, expands at the same rate as the lattice of a single crystal of the material.

In preparing the alloys for this investigation special care was given to the purity of the two constituent metals. Freedom from impurities is important with these alloys, as the presence of very small quantities of carbon is known to produce a marked effect upon the properties of nickel and iron and of their alloys. For this reason special attention was paid in the course of the work to the effect of the adhesive employed for mounting the powder on the thermo-sheet. In all the cases examined no effect could be detected, probably because the alloys were not raised to temperatures high enough to enable them to combine with the carbon. The point had however to be thoroughly investigated before it could be stated with any degree of certainty that the adhesive produced no effect.

The X-ray method of measuring the coefficients of thermal expansion of materials has been discussed in a previous paper. The accuracy of which the method is capable depends in great measure on the expansibility of the material under investigation. As a rule a fair accuracy can be attained, but what is important in the method is that only a small quantity of the material is required, and that the measurement is always made when the material is in thermal equilibrium and free from internal stresses.

In the present instance the method was found to be amply sensitive enough to establish beyond doubt the somewhat sudden changes in parameter which occur in the alloys at the temperatures of magnetic transformation. Owing to the proximity of the atomic numbers of iron and nickel, it would be impossible from the measurement of the relative intensities of the spectral lines to state whether a re-distribution of the atoms amongst the lattice points in the alloy crystals takes place in passing through the transformation temperature. Pure nickel however exhibits the same phenomenon, and it is unlikely that in this case a redistribution of atoms amongst the lattice points occurs. But there may take place, as the element or alloy passes through the transformation temperature, a reorientation of the atoms which would cause the observed abnormal increase in the expansion of the lattice followed by a sudden decrease. It would appear that at a certain temperature the thermal agitation of the atoms is sufficient to release the nickel atom from the binding forces which keep it in position, and that above this temperature it may take up any random orientation, the lattice then expanding in a uniform and normal manner.

No change in structure takes place as the alloy is taken through the temperature of

magnetic transformation. The face-centred structure persists from room-temperature through the temperature of transformation up to 606° C., the highest temperature reached in the present work. Solution of iron in nickel takes place by the substitution of an iron atom for an atom in the nickel lattice.

The results recorded in this paper show that for the alloys considered, which are rich in nickel, the addition of iron to the nickel lattice has two effects: (1) the nickel lattice is expanded approximately in proportion to the amount of iron added, and (2) the temperature of the discontinuity in the thermal-expansion curve is raised with the addition of iron, but not proportionately to the amount of iron added.

§ 7. ACKNOWLEDGMENT

The authors wish to record their indebtedness to Dr L. B. Pfeil of the Mond Nickel Company for his advice and for kindly preparing the alloys and supplying their compositions.

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THE RESISTANCE OF PALLADIUM AND PALLADIUM-GOLD ALLOYS

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ABSTRACT. Measurements of the electrical resistances of palladium and four alloys of palladium and gold have been made from liquid oxygen temperature to about 1000° C. It is found that the variation of resistance with temperature for these alloys resembles that of constantan for compositions up to about 60 atoms per cent of gold, where there is a change over to the normal type of resistance curve. This, and the results for pure palladium, are shown to be in agreement with the quantum-mechanical theory of conductivity as applied by Mott to the transition metals.

§ 1. INTRODUCTION

In two recent papers Mott[1,2] has shown that it is possible on the basis of the quantum theory developed by Bloch to explain certain anomalies in the electrical and magnetic properties of the transition metals. It is found that the energies of the electrons in a crystalline metal are confined to bands of energy levels, each band corresponding to a single stationary state of the isolated atom. The number of levels or states in each band is equal to the product of the statistical weight of the atomic state and the number of atoms in the crystal. Thus the *d* bands contain five states per atom and the *s* band one. The distribution of electrons in each band is governed by the Pauli principle; i.e. there cannot be more than two electrons in each state.

Now it is essential that if a substance is to be a conductor the bands occupied by electrons should not be completely full, for then it would be impossible for an electron to acquire energy in an applied electric field since all the states to which it might pass would be occupied. But in palladium the number of electrons per atom is just sufficient to fill all the bands up to the 4*d*. Hence the metal would be an insulator unless the 5*s* and 4*d* bands overlap, in which case the ten electrons which would otherwise fill the 4*d* band will be shared between the 5*s* and 4*d* bands and there will be vacant states in each band to which electrons may make transitions. Similarly the 4*s* and 3*d* bands of nickel and the 6*s* and 5*d* bands of platinum must overlap, and both *s* and *d* electrons make contributions to the conductivity of these metals; see figure 1.

In the classical theory of conduction the conductivity is given by $\sigma = \frac{1}{2}ne^2/m\nu$, where n is the number of electrons per unit volume, e the electronic charge, m the mass of the electron and ν the frequency with which the electron collides with and

is scattered by the atoms of the metal. In the quantum theory this formula still holds but ν must be calculated by the usual quantum-mechanical method, being proportional to the probability of scattering. Further, since we are dealing with s and d electrons in the transition metals, we must write

$$\sigma = \frac{1}{2}\frac{n_s e^2}{m_s \nu_s} + \frac{1}{2}\frac{n_d e^2}{m_d \nu_d}.$$

Calculations by Fuchs[3] show that the s electrons in copper and silver behave approximately as free electrons and the same would be expected for the s electrons of nickel and palladium which have similar atomic fields. Thus m_s is approximately the actual mass m_0 of a free electron. But the motion of the d electrons from atom to atom under the influence of an external field is much slower owing to the small overlap between one d wave function and that of a neighbouring atom. This is

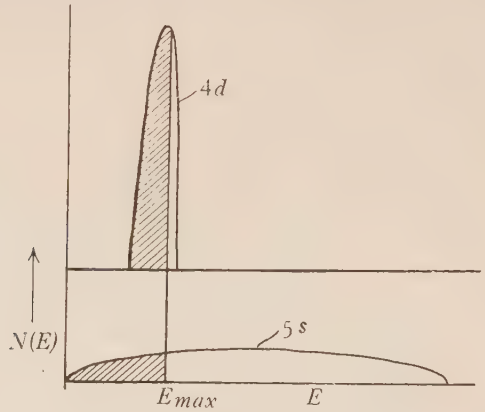

Figure 1. Density of electronic states $N(E)$ in the $5s$ and $4d$ bands of palladium plotted against their energy E. The shaded portions represent occupied states.

equivalent to taking $m_d \gg m_0$ so that the s electrons are mainly responsible for the conductivity. The conductivity thus depends on n_s and ν_s.

In a perfect lattice with no atomic vibrations the conductivity would be infinite. The scattering probability is found to be proportional to the mean-square displacement of the vibrating atoms, which is proportional to $T/M\Theta^2_D$ (where T is the absolute temperature, M is the mass of the atom, and Θ_D is the Debye characteristic temperature), and to the density of states $N(E)_{\max.}$ in the neighbourhood of the final state, i.e. at the surface of the Fermi distribution, the energy loss on scattering being small. In the normal metals copper, silver, gold, tungsten, etc., only the s electrons are conduction electrons and since they behave approximately as free electrons $N(E)$ will hardly vary with the energy of states at the surface of the Fermi distribution and the resistance is quite accurately proportional to $T/M\Theta^2_D$. Since Θ_D depends on the volume it is not constant but decreases as the metal expands. Hence the resistance actually increases more rapidly than a linear law would imply. But in the transition metals there is also the possibility of the s electrons making transitions to the d band and of d electrons making transitions to the s band as well

as of $s \rightarrow s$ and $d \rightarrow d$ transitions. Since the s electrons make the chief contribution to the conductivity we need only consider the $s \rightarrow d$ transitions.

It is essential to the theory that the d band should be narrow. This is probable owing to the small overlap between the wave functions and consequently the small interaction between the d shells of neighbouring atoms. We see, therefore, that the density of states at the surface of the Fermi distribution will be larger in the d band than in the s band. $s \rightarrow d$ transitions are therefore more probable than $s \rightarrow s$ transitions, which is the reason for the relatively large resistance of the transition metals. Moreover, a consideration of the susceptibilities of silver-palladium alloys shows that the d band is nearly full, having 9·4 electrons per atom (see below). The density of vacant states in the d band will therefore decrease rapidly with increasing energy. Hence when the metal is heated and more electrons are raised to higher states by acquisition of thermal energy, the density $N_d(E)$ of d states to which the s electrons with most energy can make transitions will have decreased and the probability of such transitions will be less. Hence as the temperature rises the electrons will remain longer in the s state before being scattered to a d state, and the resistance will increase less rapidly than that of a normal metal. For s electrons with energies greater than the upper limit of the d band this scattering-probability will be small and will decrease much less rapidly. Hence a temperature will be reached where the resistance is again approximately proportional to T, i.e. we expect a flex in the {resistance, temperature} curve. The temperature at which the flex occurs will depend on the value of $E_{\max.}$, being lower as $E_{\max.}$ increases. In addition of course the decrease of Θ_D will ultimately make the resistance increase more rapidly than the linear law as in a normal metal.

Calculation based on these considerations gives the expression for the resistance

$$\rho \propto \frac{T}{M\Theta^2_D}\left[1-\frac{\pi^2}{6}\left(\frac{T}{T_0}\right)^2\right],$$

where T_0 is a constant called the degeneracy temperature for the d shell and T is small compared with T_0. From the observed resistances, $T_0 = 2100°$ K. for palladium and 3100° K. for platinum.

Owing to the uncertainty in computing the variation of Θ_D with temperature it is simpler to express the result as

$$\left(\frac{\rho}{\rho_0}\right)=\left(\frac{\rho}{\rho_0}\right)_N(1-\text{constant } T^2),$$

where $\left(\frac{\rho}{\rho_0}\right)_N$ is the ratio for a normal metal;

or

$$\left(\frac{\rho}{\rho_0}\right)_N-\left(\frac{\rho}{\rho_0}\right)=AT^3,$$

where A is approximately constant but increases slightly with T.

By taking copper as the normal metal it is found that a formula of this type fits the experimental values for platinum quite well though the value of A actually decreases slightly with rise of temperature. It is therefore of interest to determine whether this formula also applies to palladium.

We now consider the effects of alloying palladium with metals containing one *s* electron per atom. The susceptibility of palladium-silver[4] and palladium in which hydrogen is occluded[5, 6], referred to above, decreases with increasing concentration of silver or hydrogen until, at about 60 atoms per cent, it has fallen to zero and afterwards becomes negative (diamagnetic). This is explained as follows: It is concluded that the high paramagnetism of palladium and platinum is proportional to the density of states in the *d* band, i.e. to $N_d(E)_{\max.}$. Now it is evident from figure 1 that since the density of vacant *d* states is much greater than the density of *s* states, most of the extra *s* electrons of silver or hydrogen will go into the *d* band until it is full, thus reducing the value of $N_d(E)_{\max.}$. The experiments show that about 0·6 additional electrons per atom are sufficient to fill the *d* band completely, i.e. in pure palladium there are 9·4 electrons per atom in the *d* band, the remaining 0·6 being in the *s* band.

Further evidence is obtained from the measurements of Giebel[7] on the resistance and temperature coefficient of palladium-gold alloys. The resistance of a disordered alloy is R_0+r_t at $t°$ C., where R_0 is the resistance due to disorder (*Restwiderstand*) and r_t the resistance due to thermal motion of the atoms (scattering). As gold is added to palladium we expect that the vacant *d* states will be gradually filled and consequently, owing to reduction in $N_d(E)_{\max.}$, that r_t will decrease rapidly up to about 60 atoms per cent of gold. For greater quantities of gold the increasing number of *s* electrons will cause a further decrease of resistance except that this will be counterbalanced by the greater scattering-power of the heavier gold atoms. Hence for concentrations of between 60 per cent and 100 per cent of gold, r_t will be approximately constant. The results obtained by Giebel bear out these conclusions[7].*

It has been shown above for pure palladium that as the temperature is raised and the density of *d* states to which the *s* electrons can be scattered decreases, the resistance increases less rapidly than that of a normal metal. But by the addition of gold to palladium the density of vacant *d* states is, as it were, artificially decreased. The rate of decrease of $N(E)$ with increasing E thus becomes greater and we should expect the rate of increase of r_t to be even less rapid than for palladium, except in so far as the resistance is increased by the decrease of Θ_D as gold atoms replace palladium. We should also expect that for each alloy the increase of resistance due to decrease of Θ_D with rising temperature would ultimately become predominant and that there would be a flex in the {resistance, temperature} curve as there is (theoretically) in pure palladium, and that a change over to the normal type of resistance curve would occur at about 60 per cent of gold. Experiments have therefore been made to carry the resistance-measurements for palladium to higher temperatures than heretofore and to test the above conclusions for the palladium-gold alloys.

* Cf. Rosenhall[8] for similar measurements on palladium, silver, and hydrogen. The resistance of certain palladium-silver alloys actually decreases with the addition of hydrogen owing to the rapid decrease in r_t. See figure 5 of the cited paper.

§ 2. EXPERIMENTAL METHOD

The palladium and the alloys were obtained from Messrs Johnson Matthey and Co. in the form of rods 2 mm. in diameter, the rods being 18 cm. long in the case of the palladium and 12 cm. long in the case of the alloys. The palladium was 99·98-per-cent pure and the alloys, which were specially prepared, contained respectively 25, 40, 55 and 65 atoms per cent of gold. The method used was to measure the p.d. between two points on the rod when a constant current was passed through it. The current was set to a constant value (about 2 A.) by means of a Weston substandard mirror-scale ammeter and the p.d. was measured on a Tinsley potentiometer reading in steps of 10μV. up to 19mV. The current-setting could be repeated to an accuracy greater than $\frac{1}{40}$ per cent.

The temperature was measured with a calibrated platinum, platinum-rhodium thermocouple. For the temperatures about 0° C. the specimen was mounted in a vacuum furnace consisting of a Vitreosil tube 34 in. long by $1\frac{1}{4}$ in. internal diameter wound over 15 in. of its length near the lower end with nichrome wire and containing a mild steel liner to give greater uniformity of temperature. Each rod was cut in half and the two halves R were spot-welded to nickel rods N of the same diameter and $\frac{1}{4}$ in. apart, which served as current leads, figure 2. The reason for this symmetrical arrangement is explained later. The potential leads were nickel wires attached by spot welding at P and the thermocouple was similarly attached midway between them at S. Spot welding did not seem to affect the value of ρ/ρ_0 though it usually produced an increase in ρ_0 of about 1 per cent. The whole was suspended from a glass cap C and held axially in the furnace by a quartz spacer at the lower end of the rods. The leads passed through fine tubes T in the cap; a side tube and ground joint G enabled the furnace to be connected to a diffusion pump. Sealing-wax was used for the seals, one of these near the windings having to be water-cooled. The pressure was less than 10^{-6} mm. except at the highest temperatures, where owing to the evolution of gas it sometimes rose to about 10^{-4} mm. The temperature variation along the specimen was always less than $\frac{1}{2}$° C., and thermoelectric e.m.fs. due to the temperature-gradient were eliminated in the usual way by reversing the current and potential leads. For temperatures below 0° C. the specimen was immersed in a vacuum flask containing liquid oxygen, solid carbon dioxide and acetone or liquid ethylene.

In the measurements on the 55 per cent alloy and to a less extent with the 40 per cent alloy it was observed that the p.d. increased slowly to a maximum each time the current was reversed. Tests showed that the mean value of the p.d. read immediately after reversing the current was the same as that obtained by allowing the p.d. to come to a steady value and subtracting from this the residual e.m.f. measured immediately after switching off the current. The residual e.m.f. decreased slowly after the current had been cut off. The true p.d. was therefore taken as the mean of the values immediately after reversal of the current. The effect was explained as due to Peltier heating and cooling at the junctions of the specimen and nickel rods, which in these alloys would be expected to be abnormally large. This heat

was conducted along the specimen and slowly altered the temperature at the points of contact of the potential leads. Since the effect reverses with the current, the reversal of the connections to the potential leads would make the drift in the p.d. appear in the same direction in each case. This also explains why the effect dies away when the current is cut off.

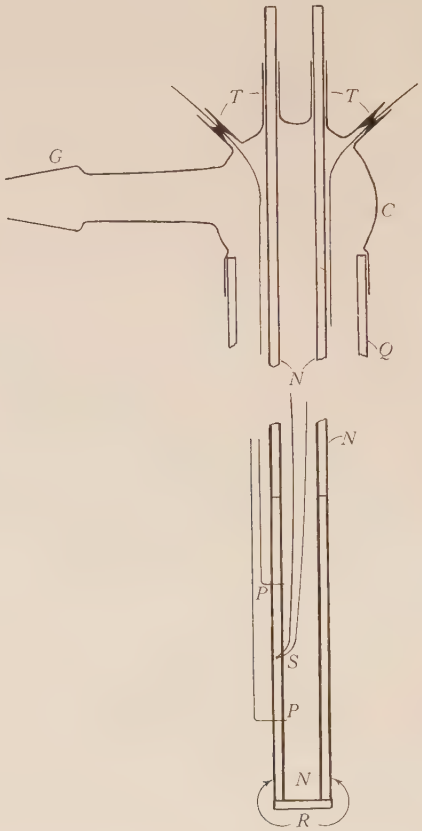

Figure 2. Symmetrical arrangement of rods R to allow for free expansion of the specimen on heating. Q is the top of the furnace tube. (Not drawn to scale.)

§ 3. RESULTS

The experimental values for palladium and the alloys are shown in table 1 as the ratio of the resistance ρ at $T°$ K. to the resistance ρ_0 at 0° C. The values for palladium agree well with those given by Holborn[9]. σ_0 is the specific resistance at 0° C.

It was observed that although the values of ρ/ρ_0 for palladium hardly changed during annealing there was a considerable increase in the specific resistance σ_0. The value of σ_0 at 0° C. after a preliminary heating to 650° C. was $10{\cdot}63 \times 10^{-6}$ ohm-cm., and after annealing at 970 to 1000° C. for three days (when the resistance appeared to have become constant) was $12{\cdot}38 \times 10^{-6}$. It was for this reason that the symmetrical arrangement, in which the specimen was allowed to expand freely

Table 1

Metal or alloy	Palladium before annealing	Palladium after annealing	65 % gold	55 % gold	40 % gold	25 % gold
σ_0 / Temp. (° K.)	$10{\cdot}63 \times 10^{-6}$	$12{\cdot}38 \times 10^{-6}$	$14{\cdot}31 \times 10^{-6}$	$20{\cdot}67 \times 10^{-6}$	$29{\cdot}46 \times 10^{-6}$	$28{\cdot}68 \times 10^{-6}$
90·2	0·2283	0·2327	0·8850	0·8901	0·9037	0·8170
169·3	0·5788	—	—	—	—	—
194·6	0·6840	—	—	—	—	—
273·2	1·000	1·000	1·000	1·000	1·000	1·000
373	1·372	1·370	1·063	1·057	1·042	1·086
473	1·730	1·708	1·128	1·116	1·080	1·166
573	2·078	2·039	1·200	1·172	1·114	1·237
673	2·395	2·357	1·271	1·221	1·145	1·299
773	2·704	2·645	1·335	1·267	1·175	1·358
873	2·972	2·907	1·403	1·314	1·204	1·416
973	3·220	3·152	1·482	1·368	1·240	1·472
1073	3·449	3·382	1·566	1·430	1·283	1·527
1173	—	3·597	1·655	1·493	1·325	1·585
1250	—	—	1·740	1·548	1·363	1·633
1273	—	3·803	—	—	—	—

during heating, was adopted. As first set up the specimen was bent slightly after being heated and it was thought that this might have produced fine cracks along the crystal boundaries. However a second specimen of palladium set up as in figure 2 also showed the same behaviour. This could not be due to the absorption of impurities given off by the iron tube, as the latter was well baked out beforehand at about 1200° C. in an induction furnace, and the palladium had not developed a *Restwiderstand*. The diameter of the specimen was measured before and after heating and the evaporation reported by Holborn[9] could not be detected. It seems possible to explain this increase by the growth of crystals in the metal and the development of small fissures between them. A similar increase was observed in the alloys and in each case examination of the specimen under the microscope after annealing revealed the presence of fairly large crystals.

The agreement between these results for palladium and the expression obtained by Mott is shown in table 2 by the constancy of the figures in the third column. Gold is chosen for the comparison, since of the normal metals its compressibility and thermal expansion are nearest to those of palladium. The values of $(\rho/\rho_0)_{Au}$ are the means of those obtained by Holborn[9] and Northrup[10].

Table 2

$(\rho/\rho_0)_{Au} - (\rho/\rho_0)_{Pd}$	T (° K.)	$\frac{\text{Column 1}}{T^3} \times 10^9$
0·181	573	9·62
0·325	673	10·66
0·469	773	10·15
1·24	1073	10·04
2·63	1273	9·84

At the highest temperatures the $\{\rho/\rho_0, T\}$ curve appears to be straightening out prior to turning upwards as was expected by the theory, that is to say the flex is outside the range of the measurements.

The values of ρ/ρ_0 for each alloy are those obtained after annealing until the resistance had become constant. In all cases the final values repeated to within 0·1 per cent.

§ 4. DISCUSSION OF RESULTS

If the resistance of each alloy is plotted against the absolute temperature, the resulting curve produced back to cut the resistance axis will give an intercept R_0 which will not differ greatly from the true *Restwiderstand*. The resistance at any

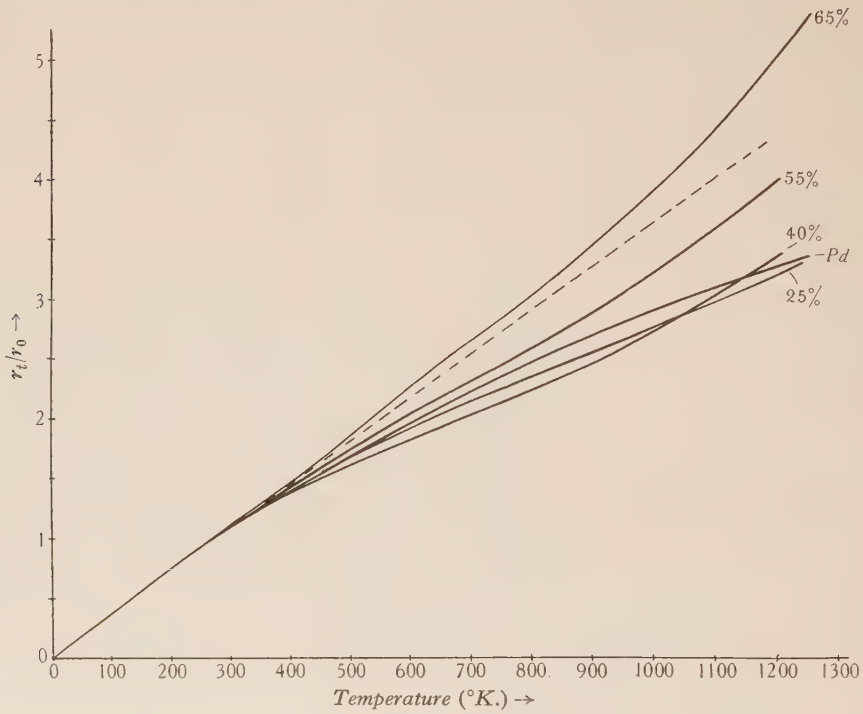

Figure 3. Relation between r_t/r_0 and the absolute temperature for pure palladium and alloys containing 25, 40, 55 and 65 atoms per cent of gold.

other temperature $t°$ C. can then be expressed as R_0+r_t. Since we are only interested in the thermal part of the resistance, the ratio r_t/r_0 is plotted against T in figure 3 for the various alloys together with the curve for pure palladium deduced in the same way.* These curves clearly show that as gold is added to palladium the resistance ratio at first increases less rapidly with increasing temperature but that a change-over occurs between 55 and 65 per cent of gold. The fact that the decrease

* Owing to the form of the {resistance, temperature} curve of palladium, which in the neighbourhood of absolute zero behaves like a normal metal, this treatment is equivalent to considering the value of R_0 for palladium to be negative.

in r_t does not continue up to 60 per cent of gold is presumably due to a decrease in Θ_D as gold is added.

It was at first expected that the position of the flex in the curve would move down to lower temperatures as the concentration of gold was increased. However, it is found that the flex temperature for the 55-per-cent alloy is intermediate between the corresponding temperatures for the 25- and 40-per-cent alloys. But the point at which the turn-up begins depends not only on the position of $E_{\max.}$ but also on the variation of Θ_D with T, and the addition of gold will probably cause changes in this variation which might account for the above expectation not being realized. It will be noticed that the variation of the resistance for the 55-per-cent gold alloy resembles that of the copper-nickel alloys containing about the same proportion of copper (constantan). The two alloys are in fact similar from a theoretical point of view though the latter is complicated by the ferromagnetism of nickel. It will be further noticed that the curve for the 65-per-cent alloy has not strictly the same shape as that for a normal metal but has a small hump at about 600° K. It seems unlikely that at this temperature there is a change in the distribution of electrons between the *s* and *d* bands and the re-appearance of some vacant states in the *d* band. It is more likely that the alloy, like Invar, has an anomalous coefficient of expansion. Thus if it contracted with rise of temperature in the region of 600° K. the consequent increase in Θ_D would cause a decrease in the resistance. There is little doubt however that in the main the results are in agreement with the theory.

§ 5. ACKNOWLEDGMENTS

The writer wishes to express his best thanks to Prof. N. F. Mott and Dr H. H. Potter for suggesting this problem, and for much helpful advice and criticism, both theoretical and experimental, throughout. He is also indebted to the Department of Scientific and Industrial Research for a personal grant and to the Colston Research Society of the University of Bristol for the provision of the alloys.

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A DETERMINATION OF THE FUNDAMENTAL TYPES OF ELECTRON OSCILLATIONS IN A TRIODE VALVE

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ABSTRACT. It is confirmed experimentally that with a retarding-field triode, electron oscillations can be maintained whose wave-lengths are determined completely by the external circuit (Gill-Morell oscillations); and that, in addition, oscillations can be obtained with wave-lengths completely independent of the external circuit, provided the latter does not approach resonance with them (Barkhausen-Kurz oscillations).

The dependence of the Barkhausen-Kurz oscillations on the valve-dimensions and operating conditions is examined in detail. It is found that for any given set of conditions there are not one, but two possible fundamental wave-lengths: type A, of shorter wave-length, and type B, of longer wave-length, both of which may give harmonics.

For any valve of grid radius r_g, and at constant anode voltage, characteristic curves

$$\frac{\lambda_A\sqrt{V_g}}{r_g}=F_A\left(\frac{i_g}{i_0}\right) \quad \text{and} \quad \frac{\lambda_B\sqrt{V_g}}{r_g}=F_B\left(\frac{i_g}{i_0}\right)$$

may be obtained, where λ_A and λ_B are the wave-lengths of the A- and B-type oscillations, V_g is the grid voltage, and i_g/i_0 is the degree of saturation of the grid-cathode space. Except for certain limiting conditions, the dependence of F_A and F_B on valve-dimensions and operating conditions is small, a slight decrease being obtained with decrease of the ratio (grid spacing)/(grid radius), or with decrease of grid voltage.

By application of the theory developed in an earlier paper, the shape of the characteristic A curve establishes that the A-type oscillations are due to the periodic transit of electrons between the cathode and virtual cathode. The shape of the characteristic B curve, on the other hand, shows that the B-type oscillations are not due to any such transit of electrons, and leads to the conclusion that they are determined by the resonant circuit formed by the grid-plate capacity in parallel with the equivalent inductance of the electron cloud between these electrodes.

§ 1. INTRODUCTION

Electron oscillations may be produced either by the retarding-field triode or by the magnetron. The latter has been examined in detail by Megaw[1]. In the present paper, only the retarding-field method is considered. Summaries of the numerous investigations employing this method have been given by Pierret[2], Wenstrom[3], Megaw[4], and Wagner and Hollmann[5].

Although electron oscillations were first discovered fifteen years ago and have since been subject to continuous investigation, there is thus far no generally accepted theory of their maintenance, and furthermore there has been considerable divergence

in the experimental results recorded. In particular, in regard to the nature of the oscillations, some workers have concluded that the wave-lengths maintained are determined completely by the external circuit (Gill-Morell oscillations), while others have found them to be determined only by the valve operating conditions and dimensions (Barkhausen-Kurz oscillations). Many other workers have found the wave-lengths to depend to some degree on all the variables.

Although the existence of Barkhausen-Kurz oscillations, as defined above, has been fairly generally accepted, a complete investigation into their dependence on conditions within the valve has not previously been attempted, most workers confining their attention to the Gill-Morell oscillations. This is probably due to the fact that, whereas the amplitude of the Gill-Morell oscillations is of the order of some milliamperes, the Barkhausen-Kurz oscillations are invariably of very small amplitude, of the order of 1 microampere. For certain critical adjustments their amplitude may increase to perhaps 10 microamperes, and such direct observations on Barkhausen-Kurz oscillations as are available have usually been confined to these optima.

With such limited results it is not possible to choose definitely between the many theories put forward to account for the Barkhausen-Kurz oscillations. These fall roughly into two classes. On the one hand are the theories which assume the oscillations to be due to some periodic transit of electrons within the valve. Various transits have been suggested, but the original theory of Barkhausen and Kurz[6], that the periodic time of the oscillations is equal to the transit time of electrons oscillating between cathode and plate, is probably most generally accepted. On the other hand are the theories which assume the oscillations to be due to a resonant circuit formed by some part of the electrode system, modified in various ways by the presence of space charge. A theory of this nature has been put forward recently by Rostagni[7].

In the present paper it is shown that in general the retarding-field triode can maintain both Gill-Morell and Barkhausen-Kurz oscillations. Particular attention has been paid to the Barkhausen-Kurz oscillations, and it has been found that there exist two distinct types: one due to the periodic to-and-fro motion of the electrons about the grid of the valve, and the other to a resonant circuit formed by the grid-plate capacity in parallel with the equivalent inductance of the electron cloud between these electrodes. The existence of two types of Barkhausen-Kurz oscillations, associated with such different mechanisms, has not hitherto been realized.

The cause of the maintenance of the oscillations is not dealt with in the present paper.

§ 2. PROCEDURE

The circuit, figure 1, used by the author is essentially the same as that used by Barkhausen and Kurz and most subsequent workers, the Lecher wires which form the tunable external circuit being connected to the grid and plate of the valve. The effective length of this circuit is altered by varying the position of the shorting condenser C_1. The grid and plate leads are connected to the terminals of this con-

denser, where there are potential nodes. The plate lead includes a suspended-coil galvanometer, of sensitivity 5.10^{-9} A./mm. at 1 m., with a variable shunt, and with provision for applying a balancing current.

The onset of oscillations is detected by the accompanying increase of plate current. It is known[8] that when the plate potential is only slightly different from that of the cathode the increase in plate current is roughly proportional to the amplitude of the oscillatory current. This method of measurement was adopted in the present experiments because of its great sensitivity. Although with all conditions adjusted for optimum maintenance oscillatory currents of the order of 5 mA. were obtained, the usual amplitudes, of less than 1 microampere, are quite impossible to measure directly. Furthermore, for accurate measurements of wave-length very slight changes in the oscillatory current must be detected.

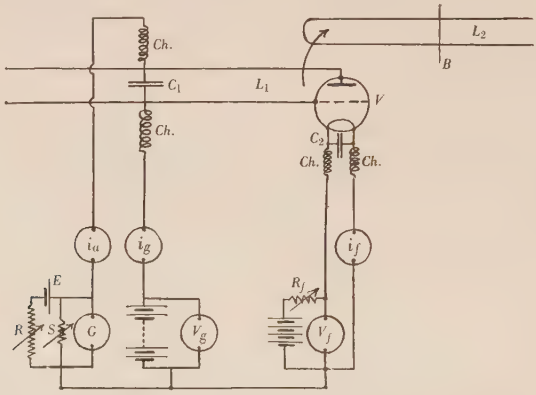


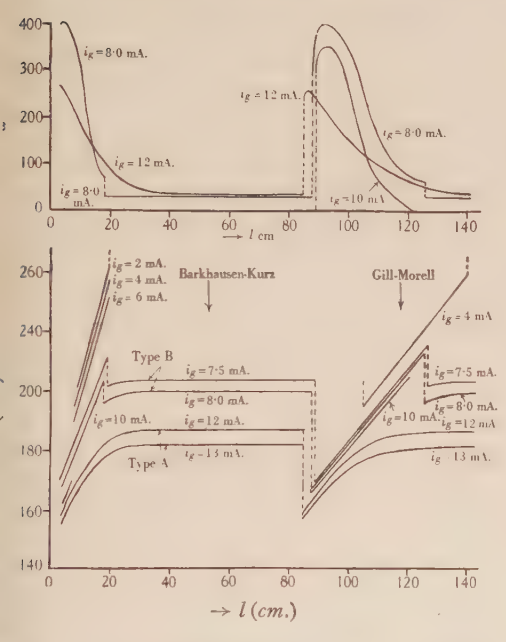
Figure 1. Circuit for the examination of electron oscillations. V, valve; L_1, external circuit; L_2, Lecher tuning circuit; B, shorting bridge; C_1, bridging condenser; C_2, filament blocking condenser; *Ch.*, chokes; S, variable shunt; E, backing e.m.f.; R, regulating resistance for E; R_f, filament current regulating resistance.

The wave-lengths of the oscillations were measured by means of the usual second Lecher-wire circuit L_2, magnetically coupled at one end to the external circuit L_1 of the valve. As their length is varied by means of the short-circuiting bridge B, the absorption of energy from the valve circuit, which occurs when the effective length of the Lechers is a multiple of half a wave-length, is indicated by a decrease of plate current.

§ 3. EFFECT OF THE EXTERNAL CIRCUIT: BARKHAUSEN-KURZ AND GILL-MORELL OSCILLATIONS

Preliminary experiments on the effect of varying the length of the external circuit showed that, for nearly all the valves examined, Barkhausen-Kurz oscillations of small amplitude could be maintained, in addition to the larger amplitude Gill-Morell oscillations. The length l of the external circuit is measured from the valve legs and does not include the portion formed by the leads inside the valve. At constant grid voltage V_g the variations of the maintained wave-length λ and of the

plate current i_a with variation of l were determined for various fixed values of grid current i_g. A typical set of results for one valve, no. 6, at $V_g = 50$ V. is shown in figure 2, which shows that between $l = 5$ cm. and $l = 20$ cm., and between $l = 80$ cm. and $l = 120$ cm., Gill-Morell oscillations are obtained whose wave-lengths are determined by the natural wave-length of the external circuit. These oscillations are maintained for any value of i_g, from 2 to 8 mA. approximately. The slight decrease in wave-length obtained with increase of i_g is due to the fact that the external



gure 2. Variation of plate current i_a and wave-length λ with circuit-length l, for various grid currents i_g. Valve 6. $V_g = 50$ V.

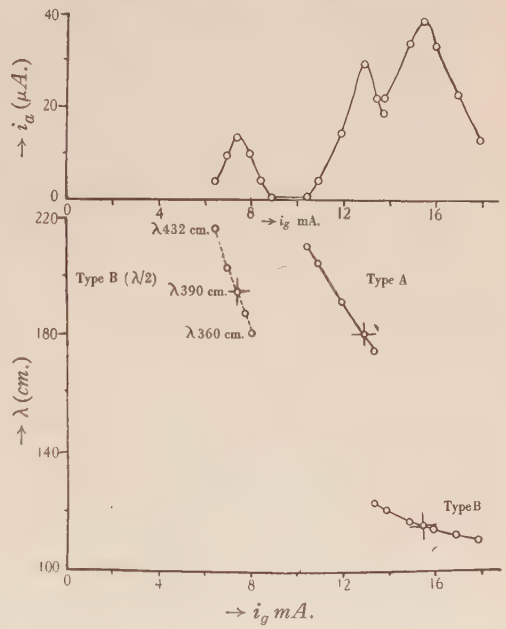


Figure 3. Barkhausen-Kurz oscillations. Variation of plate current and wave-length with grid current i_g at constant grid voltage. Valve 6. $V_g = 50$ V. $l = 30$ cm. → 70 cm.

circuit includes the grid-plate capacity; it has been established [9] that the dielectric constant of space-charge is less than unity, so that any increase in the grid current will decrease the grid-plate capacity, and hence also the natural wave-length of the external circuit.

Between $l = 30$ cm. and $l = 80$ cm. the figure shows that Barkhausen-Kurz oscillations, with wave-lengths independent of the external circuit, are maintained for values of i_g in the region of 7·5 mA. and 12 mA. respectively. No oscillations could be obtained between $i_g = 9$ mA. and $i_g = 11$ mA. It is shown later that the Barkhausen-Kurz oscillations maintained at the two values of i_g are of different types, distinguished throughout the paper as types A and B.

§ 4. THE BARKHAUSEN-KURZ OSCILLATIONS

It is clear that the wave-lengths of the Gill-Morell oscillations are determined by the characteristics of the external circuit. The following experiments are mainly concerned with the identification of the Barkhausen-Kurz oscillations.

With the external circuit maintained within such limits that only Barkhausen-Kurz oscillations were obtained, the dependence of their wave-lengths on the valve operating conditions and dimensions was examined. For this purpose a series of specially constructed valves was used in which the various dimensions were varied in turn. All the valves except one were of the AT 40 type, having concentric cylindrical electrodes, and a tungsten filament rated at 6 V., 2·5 A. The grid was a helix of wire with the turns short-circuited by the supporting bar. The pressure in these valves was of the order of 10^{-6} mm. of mercury, and was certainly not more than 10^{-5} mm. The relevant data of the valves are given in table 1.

Table 1. Data of valve-dimensions

Experiment	Valve reference	Grid radius r_g (cm.)	Anode radius / Grid radius r_p/r_g	Turns/cm. grid helix p
4·2·1	5 } AT 40	0·21	5·0	6
	6 } AT 40	0·22	3·7	6
	7 } AT 40	0·21	6·0	6
	8 } AT 40	0·21	2·95	6
4·2·2	6 } AT 40	0·22	3·7	6
	10 } AT 40	0·22	3·7	4
	11 } AT 40	0·22	3·7	8·5
	12 } AT 40	0·22	3·7	11
4·2·3	6 } AT 40	0·22	3·7	6
	17 } AT 40	0·15	3·5	6·4
	18 } AT 40	0·25	4·1	6·4
	19 } AT 40	0·35	3·5	6·4
5	20 Dario	0·15	4·1	13

AT 40 type. Tungsten filament, length 2·7 cm., radius 0·005 cm., length of grid 2·0 cm. Diameter of grid wire 0·018 cm., length of plate 1·82 cm.

Dario type. Oxide-coated filament, length 2·2 cm., radius 0·0005 cm., length of grid 1·82 cm., diameter of wires 0·018 cm., length of anode 1·5 cm.

4·1. *The dependence of the Barkhausen-Kurz oscillations on valve operating conditions.* In the first instance the valve-dimensions were maintained constant, and the dependence of the Barkhausen-Kurz oscillations on the valve operating conditions was determined. For a given valve the variation of wave-length with emission current was found for different values of grid voltage. Since in all these experiments the plate is maintained at zero potential, the plate current is extremely small, and the total emission current differs only slightly from the measured grid current.

A typical {wave-length, grid current} curve obtained with valve 6 at $V_g = 50$ V., together with the corresponding {plate current, grid current} curve, is shown in figure 3. The curves are found to be practically independent of the length l of the

external circuit between $l=30$ cm. and $l=70$ cm. It will be noted that oscillations are obtained over a wide range of grid-current values, maxima of amplitude occurring for three definite currents. It is found that with variation of grid voltage, the ratios (optimum value of grid current)/(saturation value of grid current) were practically constant. The saturation value of grid current i_0 is here defined as the value which just reduces the cathode field to zero. Actually, owing to the initial velocities of emission of the electrons, a somewhat greater current can be obtained by further increasing the cathode temperature. The experimental method adopted for measuring i_0 has been described in an earlier paper[10]. Throughout the present paper the ratio i_g/i_0 is called the "degree of saturation" and will refer to the grid-cathode space.

If for a given valve the {wave-length, grid current} curves at various grid voltages are plotted in the form $\lambda\sqrt{V_g}=f(i_g/i_0)$, they are found to correspond closely. The results for valve 6 are shown in figure 4. It is shown in § 5 that the small divergence from complete correspondence is due mainly to the slight dependence of the effective grid structure on grid voltage, and that if allowance were made for this effect the curves would correspond almost completely. The curves $\lambda\sqrt{V_g}=f(i_g/i_0)$, which are drawn for all the valves examined, will be referred to as the "characteristic curves".

From figure 4 it will be seen that there are two possible modes of oscillation, giving rise respectively to the shorter and longer Barkhausen-Kurz wave-lengths. The existence of such shorter and longer wave-lengths has been noted by Schiebe[11] and by Hollmann[12] but the matter has not been further investigated. Hollmann considered that the two modes were due primarily to the same cause, a periodic transit of electrons within the valve. The experiments described later show, on the contrary, that they are due to the oscillations of entirely different systems within the valve. They constitute two types, which are found to be of general occurrence in all the valves examined, and will be referred to as "types A and B", the shorter wave-length corresponding to type A and the longer to type B. Both types of oscillation readily give first harmonics, particularly when the wave-length of the external circuit approaches half the fundamental value. The characteristic curves are not otherwise distorted in these circumstances, and no higher harmonics have been observed. The harmonic curves have been omitted from figure 4 for the sake of clearness.

4·2. *The dependence of the Barkhausen-Kurz oscillations on valve-dimensions.* The characteristic curves provide a convenient method of showing the effect of the valve operating conditions on the wave-lengths of the Barkhausen-Kurz oscillations, and their dependence on the valve-dimensions will now be examined. Since these curves are not quite independent of grid voltage the effect of variation of valve-dimensions is shown for one voltage only, although the results in general apply at all voltages.

4·2·1. *Variation of plate radius.* Four valves, nos. 5, 6, 7, 8, were examined which differed only in their respective plate radii, the ratio of plate radius r_p to

grid radius r_g varying from $r_p/r_g = 6{\cdot}0$ to $r_p/r_g = 2{\cdot}95$. A typical set of characteristic curves obtained at $V_g = 50$ V. is shown in figure 5. It will be seen that with the exception of valve 8, which had the smallest plate radius, the characteristic curves are unaffected by variation of plate radius. A detailed discussion of the curves of valve 8 is deferred until § 5. Figure 5 also shows that for all the valves oscillations are obtainable for $i_g/i_0 > 0{\cdot}4$. This result is of some interest in view of Gill's[13] theory of maintenance, which requires that the extent of the oscillation region shall depend on plate radius.

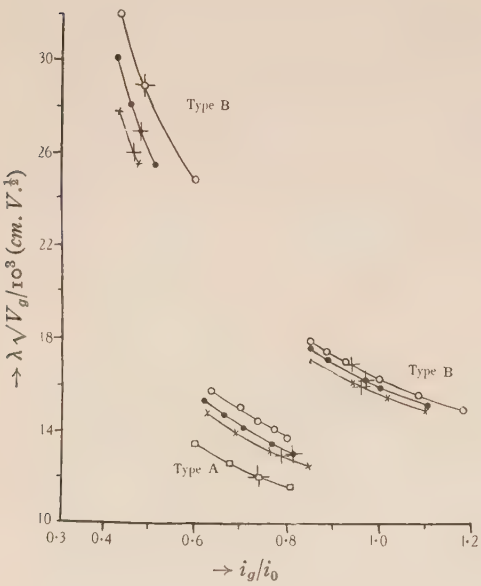


Figure 4. Characteristic curves of valve 6.
–○– $V_g = 70$ V.; –●– $V_g = 50$ V.;
–×– $V_g = 40$ V.; –□– $V_g = 30$ V.;
+ points of optimum maintenance.

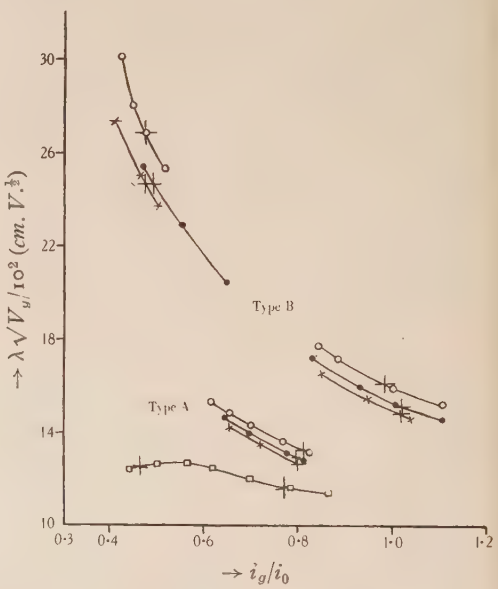


Figure 5. Dependence of characteristic curves on plate radius.
–○– valve 6, $r_p/r_g = 3{\cdot}7$; –●– valve 5, $r_p/r_g = 5{\cdot}0$;
–×– valve 7, $r_p/r_g = 6{\cdot}0$; –□– valve 8, $r_p/r_g = 2{\cdot}95$;
+ points of optimum maintenance.

4·2·2. *Variation of grid pitch.* The four valves examined, nos. 6, 10, 11, 12, differed only in the number of turns p per cm. of the grid helix, which varied from $p = 11$ to $p = 4$. Valve 10, which had a very open grid ($p = 4$), gave no detectable Barkhausen-Kurz oscillations. This is in accordance with the observations of other workers. The characteristic curves of the remaining three valves at $V_g = 50$ V. are shown in figure 6. It will be seen that the variation of grid pitch has little effect on the curves, an increase in p of 100 per cent causing a diminution of only about 10 per cent in the wave-lengths.

An interesting point arises with valve 12, which had a close-mesh grid ($p = 11$). Experiments so far have shown that the A-type oscillations are always obtained for degrees of saturation from 0·6 to 0·8, with an optimum value at about 0·8. With valve 12 this only held for $V_g < 50$ V.; for $V_g > 50$ V. the A-type oscillations are

replaced by the B type. The part of the characteristic curve obtained in this region at $V_g = 60$ V. is shown dotted in figure 6. A similar result is noted in the next section.

4·2·3. *Variation of grid radius.* In the four valves examined, nos. 6, 17, 18, 19, the grid and anode radii were varied, the ratio r_p/r_g, and all other dimensions, being maintained constant. The grid radius varied from $r_g = 0{\cdot}15$ to $r_g = 0{\cdot}35$ cm. It was found that valve 17 ($r_g = 0{\cdot}15$) gave no detectable oscillations. Taking into account the result obtained above with valve 10, it would appear that the Barkhausen-Kurz oscillations are not maintained if the ratio (grid spacing)/(grid radius) exceeds a certain value approximating to unity.

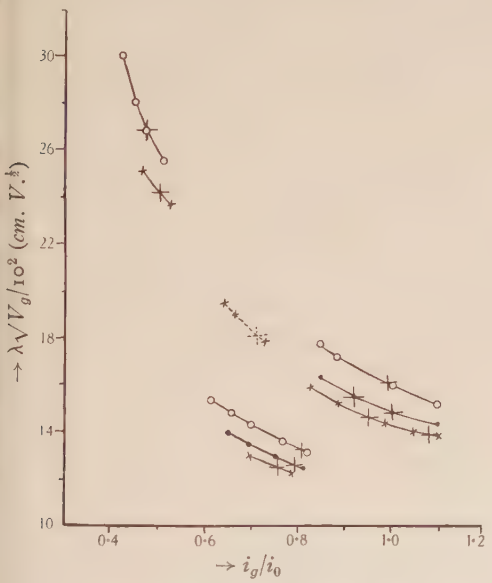


Figure 6. Dependence of the characteristic curves on grid pitch p. ($V_g = 50$ V.)

–○– valve 6, $p=6$; –●– valve 11, $p=8{\cdot}5$; –×– valve 12, $p=11$; + points of optimum maintenance; --×-- valve 12 at $V_g=60$ V.

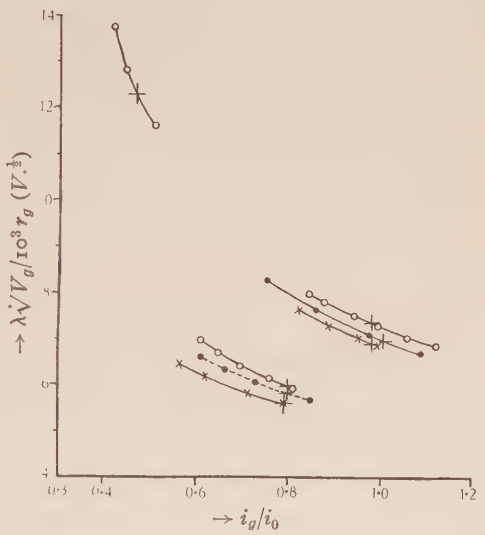


Figure 7. Dependence of reduced characteristic curves on grid radius. ($V_g = 50$ V.)

–○– valve 6, $r_g = {\cdot}22$ cm.; –●– valve 18, $r_g = {\cdot}25$; –×– valve 19, $r_g = {\cdot}35$; --●-- valve 18 at $V_g = 40$ V.

It was found that the wave-lengths of the remaining three valves were very nearly proportional to the grid radii. Reduced characteristic curves $\lambda\sqrt{V_g}/r_g = f(i_g/i_0)$ were therefore drawn for these valves. The curves obtained at $V_g = 50$ V. are shown in figure 7. It will be seen that these curves correspond closely, an increase of 50 per cent in r_g causing a diminution of 10 per cent in the values of $\lambda\sqrt{V_g}/r_g$. The cause of this variation, and of the slight variation with grid pitch and grid voltage, will be further discussed in § 5.

For valve 18, B-type oscillations replaced the A type for $V_g > 40$ V. This confirms the result obtained earlier with valve 12, that in certain cases, most probably with valves in which the ratio (grid spacing)/(grid radius) is small, increase of grid voltage tends to cause the A-type oscillations to be replaced by the B type.

4·2·4. *Summary of the results of paragraphs* 4·2. The main results with regard to the dependence of the Barkhausen-Kurz oscillations on the valve-dimensions may be briefly summarized as follows. For nearly all the valves examined Barkhausen-Kurz oscillations are obtained for any value of $i_g/i_0 > 0{\cdot}4$, provided the ratio (grid spacing)/(grid radius) < 1 approximately. These oscillations are of two types: type B, of longer wave-length, are usually most strongly maintained at $i_g/i_0 = 0{\cdot}5$ and $i_g/i_0 = 1{\cdot}0$; and type A, of shorter wave-length, at $i_g/i_0 = 0{\cdot}8$, although with increase of grid voltage type A are liable to be replaced by type B.

The reduced characteristic curves $\lambda_A \sqrt{V_g}/r_g = F_A(i_g/i_0)$ and $\lambda_B \sqrt{V_g}/r_g = F_B(i_g/i_0)$ are, to a first approximation, independent of valve-dimensions, if $r_p/r_g > 3$. These curves afford, therefore, a most convenient way of representing the results obtained with any valve. Since they do depend to some extent, however, on the grid voltage and on the grid structure, it will be necessary, until the cause of this dependence has been examined more thoroughly, to specify these conditions when giving the characteristic curves.

When $r_p/r_g < 3$, F_A is subject to modification; this effect is fully discussed in the next section.

§ 5. THE NATURE OF THE BARKHAUSEN-KURZ OSCILLATIONS: TYPE A

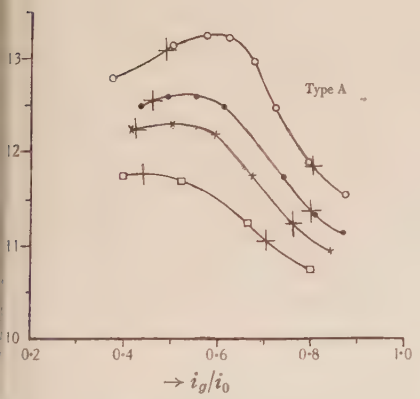
The nature of the Barkhausen-Kurz oscillations will now be considered. The existence of reduced characteristic curves which are practically independent of valve-dimensions is consistent with most of the theories which have been put forward to account for these oscillations. Fortunately definite evidence of the nature of the A-type oscillations can be obtained from the characteristic curves of two of the valves examined, nos. 8 and 20, which are shown in figures 8 and 9. For both these valves the A-type oscillations extend over a much greater range than is usual, the B type only occurring for $i_g/i_0 > 1$, and in the case of valve 8 only when $V_g > 70$ V.

The particular interest in the curves lies in their unusual shape. Referring to figure 8, it will be seen that as i_g/i_0 is increased, the characteristic A curves show an increase in wave-length preceding the usual decrease which has been observed in all the other valves. It will now be shown that this effect can only be explained on the view that the periodic time of the A-type oscillations is determined by the time of transit of electrons within the valve, and that part of this transit occurs in the grid-plate space.

It is first necessary to examine the conditions obtaining between the grid and plate. Of the electrons leaving the cathode, only a certain proportion strike the grid immediately, the remainder penetrating into the grid-plate space. Here they experience a retarding field which brings them to rest at or near the plate. They then accelerate back towards the grid where again a certain proportion pass through, to be brought to rest at the cathode, when the process is repeated. The current passing between the grid and plate is thus a definite fraction of the total emission current, depending on the proportion captured by the grid at each transit. For

some value of emission current, the current passing between the grid and plate will be sufficient to reduce the field at the plate to zero, just as in a normal diode, and the space is then saturated. Of course this can only happen if the grid-cathode space does not itself become saturated first.

Gill[13] has shown that the onset of grid-plate saturation, when the plate is a few volts positive to the cathode, may be detected by measuring the dependence of the plate current on emission current. By extrapolating the results obtained at various plate potentials an approximate result for plate at zero potential is obtained. In the case of valve 8 it was found that at any given grid voltage the onset of grid-plate saturation corresponded with the maxima in wave-length shown in figure 8. For all the other valves, owing to their larger plate radii, grid-plate saturation set in at much lower values of i_g/i_0. Since oscillations have not been obtained for such low values, the maxima in wave-length are not observed with these valves.



ure 8. Characteristic curves of valve 8. $r_g = 2{\cdot}96$.)–○–$V_g = 70$ V.; –●–$V_g = 50$ V.; – $V_g = 40$ V.; –□– $V_g = 30$ V.; + points of imum maintenance.

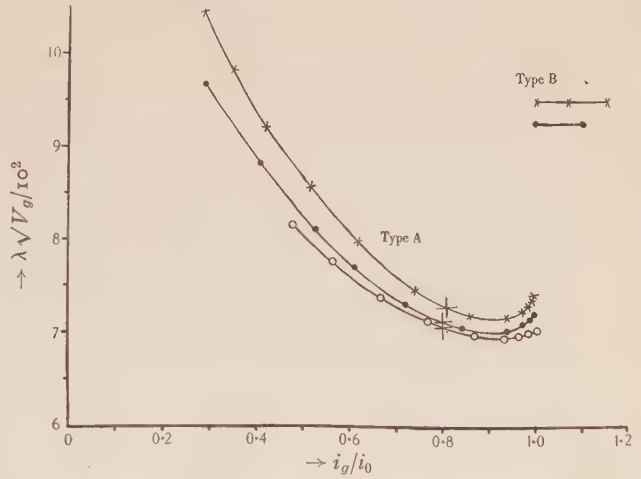


Figure 9. Characteristic curves of valve 20.
○—○– $V_g = 20$ V.; –●—●– $V_g = 30$ V.; –×—×– $V_g = 40$ V.

It has been shown theoretically in an earlier paper[10] that the time of transit of electrons in a diode increases with increasing degree of saturation, most of the increase occurring just before the onset of saturation; and that this increase at saturation becomes more apparent as the anode voltage is increased. Although the theory was developed for plane electrodes it will hold qualitatively for cylindrical electrodes. Applying these results to the grid-plate space of the valve, where the grid forms the anode with a virtual cathode at the plate, it will be seen that the increase in wave-length, and hence of periodic time of the oscillations, corresponds with the increase of electron transit time between grid and plate at the onset of saturation of the space; and, further, that the increase in wave-length becomes more apparent as the grid voltage is increased. Since such an increase is deducible on no other theory, the effect must be regarded as establishing definitely that the periodic time of the A-type oscillations is determined by the time of transit of

electrons within the valve, and that part of the transit occurs between grid and plate.

With regard to the subsequent decrease in wave-length observed with increase of i_g/i_0, it has been pointed out by Gill[13] and by Tonks[14] that with any further increase in current the virtual cathode must move away from the plate towards the grid. This causes the electron transit time between grid and virtual cathode to decrease, producing the decrease in wave-length normally observed with increase of emission current.

Referring again to figure 8, it may be noted that the onset of grid-plate saturation, indicated by the maximum in wave-length, occurs at greater values of i_g/i_0 as V_g is increased. Experiments in which the plate was used as collecting electrode for those electrons which escaped capture by the grid showed that this variation with grid voltage was due partly to the variation of the grid-capture fraction, and partly to the variation of the effective length of the grid-plate space. If the effective grid structure were independent of the grid voltage, the maxima would occur at a constant value of i_g/i_0, and if the abscissae of the curves in figure 8 were adjusted in the correct proportion for this condition to be simulated, the curves would correspond closely except in the region of the maxima. Here a variation with grid voltage is to be expected, since the increase of transit time, and hence of wave-length, becomes less with increase of grid voltage; and, further, the velocity distribution of the electrons will tend to round off the cusp that would otherwise be expected. If the same adjustments were made to the curves of the other valves, nos. 5, 6, and 7, having the same grid structure as valve 8, a similar correspondence would be obtained. Thus the dependence of the characteristic curves on grid voltage is due mainly to the variation of the effective grid structure. As is shown in the Appendix, a further slight variation in the curves is also to be expected on account of the finite velocities of emission of the electrons from the cathode.

Further observations on the onset of grid-plate saturation showed that the value of i_g/i_0 at which this occurs decreases slightly with increase of grid radius, or of the number of turns per cm. of the grid helix. This explains the slight decrease in wave-length observed in these circumstances (paragraphs 4·2·2 and 4·2·3).

The main conclusions drawn from the curves of valve 8 with regard to the nature of the A-type oscillations are corroborated and extended by the curves of valve 20. This valve is of similar type to the others which have been examined, but differs in an important particular. Whereas all the other valves had high-temperature tungsten cathodes, valve 20 had a low-temperature oxide-coated cathode. Referring to figure 9 it will be seen that at $i_g/i_0 = 1$, the characteristic curves show a small but quite definite rise, which becomes more apparent as the grid voltage is increased. It is clear that this increase in wave-length is attributable to the increase in the time of transit of the electrons between cathode and grid. The effect is small, since the cathode-grid transit time is normally only a small part of the total transit time which determines the periodic time of the A-type oscillations. It is not observed with any of the other valves, since they have high-temperature cathodes, and the electrons have a consequent large average emission velocity. It has been

shown theoretically[10] that this will have the effect of making the increase of transit time at saturation less apparent.

The qualitative results obtained with valves 8 and 20 thus establish that the A-type oscillations consist of a periodic transit of electrons which is affected by the conditions of both cathode-grid and grid-plate spaces. The most probable and only feasible electron path is from the cathode through the grid to the position of zero potential in front of the plate, and back to the cathode. This is indeed the original hypothesis put forward by Barkhausen and Kurz to explain the oscillations first observed by them. Assuming that the periodic time of the A-type oscillations is equal to twice the time of transit of an electron from cathode to virtual cathode, it is possible to calculate exactly the part of the characteristic curve obtained after grid-plate saturation has set in, provided certain data regarding the effective grid structure are known. In the case of valves 5, 6, 7, 8, these data may be obtained from the curves of valve 8. The calculation is given in Appendix (1); and in figure 10 the theoretical curve thus obtained for $V_g = 50$ V. is shown in comparison with the corresponding practical curves of valves 5, 6, 7, 8.

The quantitative agreement of the curves of valves 5, 6, and 7 with the theoretical curve is good. In the case of valve 8 it will be seen that as i_g/i_0 tends to unity the wave-lengths predicted by the theory are too small; this is due to neglecting in the calculations the increase in cathode-grid transit time which occurs at this stage. For the lower values of i_g/i_0, in the region of the maximum, there is considerable discrepancy between the theoretical curve and that of valve 8. This is due to the distribution of electron velocities, which although always obtained in practice was not considered in the calculations. As has been pointed out earlier, the distribution of velocities rounds off the cusp which would otherwise be expected at the maximum, and it is evident that the effect is large and extends considerably on either side of this point. In this connexion it is interesting to note that if the plate radius of valve 8 were still further reduced, the part of the curve where the wave-length decreases with increasing current would diminish in extent. Eventually, when $r_p/r_g = 2{\cdot}5$ approximately, saturation of the grid-plate and cathode-grid spaces would occur together, and the wave-length would then be practically independent of i_g/i_0. This has been confirmed experimentally in the case of an MT 5 valve in which $r_p/r_g = 2{\cdot}5$.

§ 6. THE NATURE OF THE BARKHAUSEN-KURZ OSCILLATIONS: TYPE B

Now that the mechanism of the A-type oscillations is established, it becomes clear that the nature of the B-type oscillations is entirely different. In the first place, if the A-type oscillations are determined by the transit of electrons from cathode to virtual cathode and back, it is difficult to visualize a longer periodic transit which would give rise to the longer B-type. Again, the effect of the external circuit is markedly different. Referring to figure 2 it will be seen that as the external circuit is shortened and approaches resonance with the A-type oscillations maintained at

$i_g = 12$ mA., their wave-length is gradually pulled into tune with that of the external circuit, this being accompanied by a large increase in amplitude. This pulling-into-tune effect was examined by Hollmann[12], who showed that it was due to the shortening of the electron transit time by the increasing amplitude of the oscillatory voltage on the grid. If the B-type oscillations were also determined by an electron transit, a similar pulling-into-tune effect would be expected in their case also. It is significant that this is not observed; the B-type oscillations are simply replaced by the true external circuit oscillations.

Careful observation of any of the characteristic B curves fails to reveal any increase in wave-length at the onset of cathode-grid saturation, such as was observed in the A curve of valve 20 (figure 9). As regards the more obvious effect, the

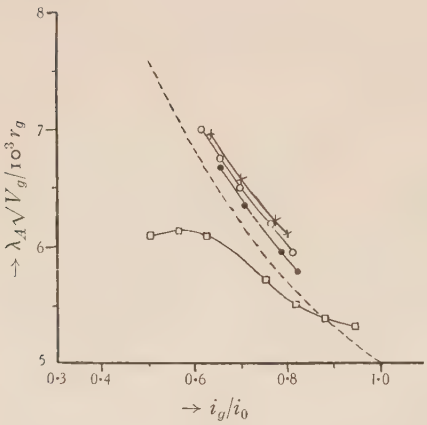


Figure 10. Comparison of theoretical and experimental characteristic A curves. ($V_g = 50$ V.)

–×– valve 5; –○– valve 6; –●– valve 7; –□– valve 8; ---- theoretical curve.

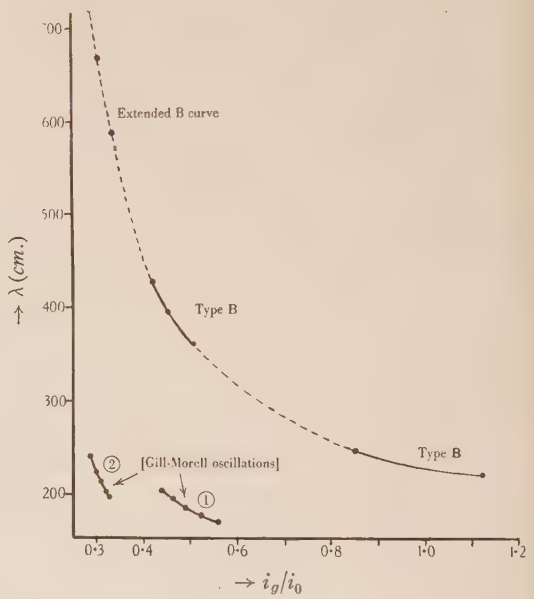


Figure 11. Extension of the characteristic B curve from observations on Gill-Morell oscillations. (Valve 6, $V_g = 50$ V.)

maximum in wave-length occurring at the onset of grid-plate saturation, direct observation has not been possible, since no valve has yet been found to give B-type oscillations at this point. With several valves oscillations were observed to commence at degrees of saturation only slightly greater. Thus, in the case of valve 6, it may be calculated from the dimensions and the observations on valve 8, that grid-plate saturation occurs at $i_g/i_0 = 0{\cdot}3$ approximately, for $V_g = 50$ V. The B-type oscillations commence at $i_g/i_0 = 0{\cdot}4$, but there is no sign of any limitation of wave-length.

However, from observations on the true external circuit (Gill-Morell) oscillations, the B curve can be extended to lower values of i_g/i_0, and, as is shown below, this extension makes it quite clear that the B-type oscillations are not determined by transit time.

Extension of the characteristic B curve by means of observations on the Gill-Morell oscillations. An examination of the Gill-Morell oscillations obtained with any given valve showed that there was, for each wave-length maintained, a definite value of i_g/i_0, nearly independent of grid voltage, at which maximum amplitude of oscillations was obtained. In figure 11 are shown the results obtained with valve 6 at $V_g = 50$ V., the wave-length being shown as a function of the corresponding optimum degree of saturation. For comparison the corresponding B curve is shown. It will be seen that the results obtained with the Gill-Morell oscillations give two distinct curves marked (1) and (2); and that for curve (1) the wave-lengths are exactly half the corresponding B wave-lengths. It is clear, therefore, that in this region the value of i_g/i_0 at which the Gill-Morell oscillations are most strongly maintained occurs when the first harmonic of the B oscillations is in resonance with them. It is reasonable to assume that in the region of curve (2) optimum maintenance of the Gill-Morell oscillations occurs when their wave-length corresponds with the second harmonic of the B-type oscillations. The corresponding portion of the B curve obtained on this assumption is shown by the broken line in figure 11. It will be seen that the parts of the B curve obtained directly, and the extension deduced from the Gill-Morell observations, form a smooth curve, thus confirming the deduction.

The characteristic B curve of valve 6, figure 12, has now been extended down to $i_g/i_0 = 0{\cdot}3$ at which point saturation of the grid-plate space is just occurring. It is clear that there is no limitation of wave-length, and that as $i_g/i_0 \to 0$ the wave-length tends to infinity. There is thus, for the B-type oscillations, a complete absence of any of the effects observed with those of the A type, and it may be taken as established beyond doubt that they are not determined by a periodic transit of electrons.

The only theory that appears capable of explaining the observed results is that of Rostagni[7], who from observations on the Gill-Morell oscillations infers the existence within the valve of a resonant circuit formed by the electron cloud between the grid and plate. He has shown that a system of electrons in statistical equilibrium between a pair of electrodes will, owing to its inertia, behave like an inductance when an a.c. potential is applied to the electrodes; this inductance in parallel with the electrode capacity forming a resonant circuit. Rostagni shows that for a uniform distribution of electrons between plane parallel electrodes, the natural pulsatance ω of this circuit is given by

$$\omega = \sqrt{\frac{4\pi N e^2}{m}},$$

where N is the number of electrons per cm^3 and m and e have their usual meanings. According to this formula the wave-length of the system varies inversely as the square root of the electron-density, and the theory will therefore yield a characteristic curve similar to that obtained experimentally.

A possible criticism of the application of the theory is the apparent non-uniformity of the electron-density within the valve, since it is difficult to visualize how

a non-uniform system can have one definite frequency. There are, however, two main causes of non-uniformity. In a normal diode the electron-density will be greatest at the cathode and least at the anode; on the other hand, with a current passing between cylindrical electrodes, the electron-density will tend to be greater at the smaller electrode. Thus in the grid-plate space of the retarding field triode, where a virtual cathode is formed at or near the larger electrode, we may reasonably expect the electron-density to be fairly constant.

In appendix (2), Rostagni's theory is applied to the calculation of the characteristic B curve, but as might be anticipated the result is very approximate. The theoretical curve obtained for valve 6 at $V_g = 50$ V. is shown in figure 12. It will be seen that the observed wave-lengths are longer than those predicted by the theory.

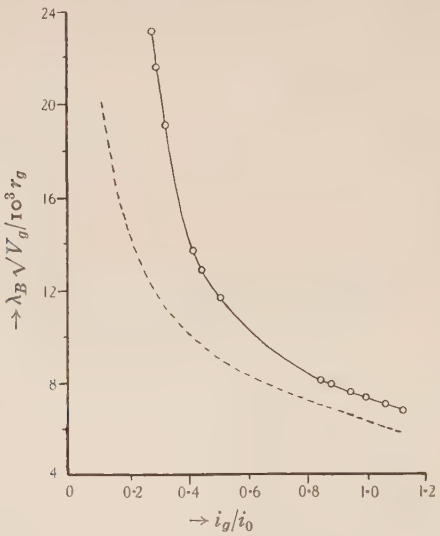


Figure 12. Comparison between theoretical and practical characteristic B curves. (Valve 6, $V_g = 50$ V.) - - - theoretical curve; — practical curve.

This may reasonably be ascribed, in part, to the increase of the electrode capacity due to their supports, an effect which was not allowed for in the calculations. There is a further cause for the considerable divergence which occurs as i_g/i_0 is reduced below 0·4. As is clear from the curve of valve 8, figure 10, a shortening in the transit time from the calculated value may occur considerably before the grid-plate space becomes unsaturated. This will produce a decrease in the electron-density, and so cause the B wave-lengths to be longer than the calculated value.

§ 7. CONCLUSION

It has been shown that a cylindrical triode with positive grid, and with plate at a potential near that of the cathode, will maintain oscillations whose periodic time is comparable with the time of transit of electrons within the valve, provided that the ratio (grid spacing)/(grid radius) is not greater than a certain value which approxi-

mates to unity. If the external circuit presents a wave-length suitable for maintenance, oscillations will be maintained in the external circuit (Gill-Morell oscillations); otherwise oscillations of much smaller amplitude are maintained within the valve, their wave-length being determined by the valve-operating conditions and dimensions (Barkhausen-Kurz oscillations).

The Barkhausen-Kurz oscillations are of two types: a shorter type A due to the periodic transit of electrons from the cathode to the virtual cathode and back, and a longer type B due to a resonant circuit formed by the grid-plate capacity in parallel with the equivalent inductance of the electron cloud between these electrodes. The type actually obtained depends mainly on the degree of saturation of the grid-cathode space. The reason for this is not so far apparent.

For every valve, characteristic curves

$$\frac{\lambda_A\sqrt{V_g}}{r_g}=F_A\left(\frac{i_g}{i_0}\right) \quad \text{and} \quad \frac{\lambda_B\sqrt{V_g}}{r_g}=F_B\left(\frac{i_g}{i_0}\right)$$

may be obtained, and over a wide range of values of i_g/i_0, F_A and F_B are nearly independent of valve-dimensions. They decrease slightly with decrease of grid voltage, or with decrease of the ratio (grid spacing)/(grid radius). Variation of plate radius has no effect on the extent of the curves, but when $r_p/r_g<3$, F_A is subject to modification, and with further reduction of r_p/r_g, $F_A\,(i_g/i_0)$ becomes practically constant.

Some of the data obtained in the course of this investigation bear closely on the cause of maintenance of the oscillations. It is hoped to deal with this aspect of the problem in a future communication.

§ 8. ACKNOWLEDGMENT

I should like to thank Dr D. Owen for his very helpful criticism and interest during the preparation of this paper.

APPENDIX

§ 1. CALCULATION OF THE CHARACTERISTIC A CURVE AFTER GRID-PLATE SATURATION HAS OCCURRED

Considering a valve with concentric cylindrical electrodes, let r_c, r_g, and r_p be the radii of the cathode grid and plate respectively; let the grid be maintained at a positive potential V_g, and the plate at zero potential relative to the cathode. Referring to figure 13, let i_g be any current in the grid-cathode space and i_u be the corresponding current in the grid-plate space, such that a virtual cathode is formed of radius $r_u<r_p$. It is assumed in the first instance that electrons are emitted from the cathode with negligible initial velocities.

Owing to grid capture, i_u will not be equal to i_g but to some fraction ψ of it. It is assumed that ψ is constant with variation of current. Thus

$$i_u = \psi i_g .$$

Then using Langmuir's[15] equation for the saturation current between cylindrical electrodes,

$$i_u = \psi i_g = \frac{2\sqrt{2}}{9}\sqrt{\left(\frac{e}{m}\right)}\kappa\frac{L_1 V_g^{\frac{3}{2}}}{\beta_u^2 r_g} \qquad \text{.....(1)}$$

$$= 14{\cdot}65 \cdot 10^{-6}\frac{L_1 V_g^{\frac{3}{2}}}{\beta_u^2 r_g}\text{ampères} \qquad \text{.....(1}a\text{)},$$

where e and m have their usual meanings, κ is the dielectric constant of the medium, L_1 the effective length of the electrodes in the grid-plate space, and $\beta_u^2 = f(r_u/r_g)$. The variation of β_u^2 with r_u/r_g is shown in figure 14.

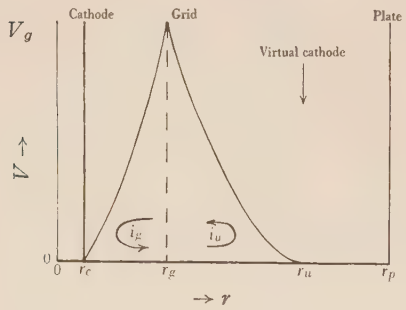


Figure 13. Voltage distribution after saturation of the grid-plate space has set in.

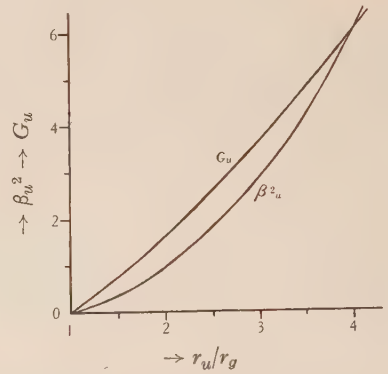


Figure 14. Variation of G_u and β_u^2 with r_u/r_g.

Similarly the grid-cathode saturation current i_0 is given by the equation

$$i_0 = 14{\cdot}65 \cdot 10^{-6}\frac{L_2 V_g^{\frac{3}{2}}}{\beta_c^2 r_g}\text{ ampères} \qquad \text{.....(2)},$$

where L_2 is the effective length of the electrodes in the grid-cathode space, and $\beta_c^2 = f(r_c/r_g) = 1{\cdot}1$ for the values of r_c/r_g such as occurred in the experiments.

Then from equations (1 a) and (2),

$$\beta_u^2 = 1{\cdot}1\frac{L_1}{\psi L_2}\frac{1}{i_g/i_0} \qquad \text{.....(3)}.$$

Now since for the valves used in the experiments the lengths of the cathode and plate were maintained constant, the ratio L_1/L_2 depends only on the grid structure; and equation (3) shows that β_u^2, and hence r_u, is determined by this structure, and by the degree of saturation of the grid-cathode space. The position of the virtual cathode is thus independent of the plate radius. The function $L_1/\psi L_2$ cannot as yet be calculated directly from the dimensions of the grid. However, for one particular value of $i_g/i_0 = i_p/i_0$ the virtual cathode will be formed at the plate, when $r_u = r_p$,

and the corresponding value of $\beta_u^2 = \beta_p^2$ is then known. In the case of valve 8 the value of i_p/i_0 is indicated definitely by the maximum in wave-length, and since

$$1{\cdot}1\,\frac{L_1}{\psi L_2} = \beta_p^2\,\frac{i_p}{i_0} \qquad \text{.....(3}a\text{)},$$

the function $1{\cdot}1\ L_1/\psi L_2$ may be calculated. The value obtained will, of course, apply also to the valves 5, 6, and 7, which have the same grid structure as valve 8; and for these valves the position of the virtual cathode can be calculated from equation (3) for any value of i_g/i_0.

The electron transit time T_{GU} between grid and virtual cathode is given by McPetrie's[16] equation

$$T_{GU} = G_u \frac{2r_g}{\sqrt{(2V_g\, e/m)}} \qquad \text{.....(4)},$$

where $G_u = f(r_u/r_g)$ and is obtained from figure 14.*

The cathode-grid transit time T_{CG}, when a negligible current is passing, is calculated from Schiebe's[11] equation

$$T_{CG} = f\left(\frac{r_c}{r_g}\right) \frac{2r_g}{\sqrt{(2V_g\, e/m)}} \qquad \text{.....(5)},$$

where $f(r_c/r_g)$ approximates to 0·6 for the usual values of r_c/r_g such as obtained in the experiments. In order to simplify the ensuing calculations, it is assumed that the cathode-grid transit time remains constant, the increase with current being neglected; this will affect the final result only slightly since T_{CG} is normally small compared with T_{GU}.

So far the effect of the initial velocities of emission has not been considered. The main effect is to reduce the transit time from the value for zero initial velocities, and this effect is normally too large to be neglected. It has been shown[10], in the case of plane electrodes carrying their saturation current, that the transit time when the electrons are emitted with an average initial velocity of V_0 volts is approximately $\{1 - 2\,(V_0/V_g)^{\frac{1}{2}}\}$ times the transit time when $V_0 = 0$. Assuming this correction to apply in the present case, the periodic time T_A of the oscillations is given by

$$T_A = 2\,(T_{GU} + T_{CG})\,\delta \qquad \text{.....(6)},$$

where

$$\delta = 1 - 2\,(V_0/V_g)^{\frac{1}{2}} \qquad \text{.....(7)};$$

and from equations (4) and (5)

$$\lambda_A = cT_A = \frac{2cr_g}{\sqrt{(2V_g\, e/m)}}\{2G_u + 1{\cdot}2\}\,\delta,$$

where c is the velocity of the electromagnetic waves. Substituting numerical values for e, m, c, and with λ_A and r_g in cm. and V_g in volts,

$$\frac{\lambda_A \sqrt{V_g}}{r_g\ 10^3} = F\,(i_g/i_0)\,\delta \qquad \text{.....(8)},$$

where

$$F\,(i_g/i_0) = 2G_u + 1{\cdot}2 \qquad \text{.....(9)}.$$

It will be seen that owing to the finite velocities of emission the characteristic curve

* A direct method of calculating T_{GU} has been given recently by Fortescue[17].

given by equations (8) and (9) will not be completely independent of V_g. In the following table, $F(i_g/i_0)\,\delta$ is calculated for valves 5, 6, 7, 8, for a range of values of i_g/i_0 at $V_g = 50$ V.

For valve 8 in which $r_p/r_g = 2{\cdot}95$, $\beta_p^2 = 2{\cdot}8$; and by experiment (figure 8), $i_p/i_0 = 0{\cdot}54$ at $V_g = 50$ V.; thus from equation (3*a*)

$$1{\cdot}1\,\frac{L_1}{\psi L_2} = 1{\cdot}52,$$

and equation (3) becomes

$$\beta_u^2 = \frac{1{\cdot}52}{i_g/i_0}.$$

For the cylindrical tungsten cathodes such as were used in the experiments $V_0 = 0{\cdot}2$ V. approximately, and

$$\delta = 1 - 2\,(0{\cdot}2/50)^{\frac{1}{2}} = 0{\cdot}87.$$

i_g/i_0	β_u^2	r_u/r_g	G_u	$F(i_g/i_0)$	$F(i_g/i_0)\,\delta$
0·5	3·02	3·05	3·75	8·7	7·55
0·6	2·52	2·83	3·30	7·8	6·8
0·7	2·15	2·66	2·95	7·1	6·2
0·8	1·89	2·55	2·70	6·6	5·7
0·9	1·68	2·42	2·45	6·1	5·3
1·0	1·52	2·32	2·27	5·74	5·0

The curve $\lambda_A \sqrt{V_g}/10^3\, r_g = F(i_g/i_0)\,\delta$ is shown in figure 10.

§ 2. CALCULATION OF THE CHARACTERISTIC B CURVE

2·1. *Derivation of Rostagni's equation.* Rostagni's equation may be derived most simply in the following manner. For a fuller treatment the original paper[7] should be consulted.

Consider a pair of plane parallel electrodes of area A, separated a distance d, with a cloud of electrons of uniform density N in statistical equilibrium between them. Let a small a.c. potential $\overline{V} = V_0\,\epsilon^{j\omega t}$ be applied between the electrodes.

Then if we neglect collisional friction

$$m\,\frac{d^2x}{dt^2} = -e\,\frac{V_0}{d}\,\epsilon^{j\omega t} \qquad \text{.....(10)},$$

whence integrating

$$\frac{dx}{dt} = \frac{j}{\omega}\,\frac{e}{m}\,\frac{V_0}{d}\,\epsilon^{j\omega t} \qquad \text{.....(11)},$$

the constant of integration being zero, since the electrons are in statistical equilibrium

The a.c. current $\bar{\imath}$ between the electrodes is given by

$$\bar{\imath} = -AeN\,\frac{dx}{dt} = -\frac{j}{\omega}\,\frac{Ne^2}{m}\,\frac{A}{d}\,V_0\,\epsilon^{j\omega t} \qquad \text{.....(12)},$$

$$= \overline{V}/j\omega L,$$

where $$L=\frac{m}{Ne^2}\frac{d}{A} \qquad \text{.....(13),}$$

the cloud of electrons thus corresponding to an inductance.

The capacity C of the electrodes is given by

$$C=\frac{A\kappa}{4\pi d} \qquad \text{.....(14).}$$

Thus the system has a natural pulsatance

$$\omega=\frac{1}{\sqrt{CL}}=\sqrt{\frac{4\pi Ne^2}{\kappa m}} \qquad \text{.....(15),}$$

and if λ_B be the corresponding wave-length

$$\lambda_B=\frac{c}{e}\sqrt{\frac{\pi m\kappa}{N}},$$

or substituting numerical values

$$\lambda_B=\frac{3{\cdot}35\cdot 10^6}{\sqrt{N}}\text{ cm.} \qquad \text{.....(16).}$$

2·2. *Calculation of the average value of N.* Referring to figure 13, if n be the total number of electrons present in the grid-plate space

$$ne=i_u T_{GU};$$

but the volume of the space containing the electrons is given by

$$v=\pi\,(r_u^2-r_g^2)\,L_1,$$

whence $$N=\frac{n}{v}=\frac{i_u}{\pi e}\frac{T_{GU}}{(r_u^2-r_g^2)\,L_1} \qquad \text{.....(17).}$$

Substituting for i_u, β_u^2, and T_{GU} from equations (1), (3) and (4),

$$N=\frac{4\kappa}{9\pi e}\,\frac{G_u\,(i_g/i_0)\,\delta}{\{r_u^2/r_g^2-1\}\,\{1{\cdot}1\,L_1/\psi L_2\}}\,\frac{V_g}{r_g^2} \qquad \text{.....(18),}$$

the correction δ (equation 7) having been inserted since $V_0\neq 0$.

With V_g in volts and r_g in cm., $4\kappa/9\pi e=10^6$; hence substituting for N in equation (16) and rearranging

$$\frac{\lambda_B\sqrt{V_g}}{10^3 r_g}=F_B\left(\frac{i_g}{i_0}\right)\frac{1}{\sqrt{\delta}} \qquad \text{.....(19),}$$

where $$F_B\left(\frac{i_g}{i_0}\right)=3{\cdot}35\sqrt{\frac{1{\cdot}1\,L_1/\psi L_2\,\{r_u^2/r_g^2-1\}}{(i_g/i_0)\,G_u}} \qquad \text{.....(19}a\text{).}$$

Substituting the results obtained earlier for valves 5, 6, 7 and 8,

$$1{\cdot}1\,L_1/\psi L_2=1{\cdot}52,\quad \delta=0{\cdot}87,$$

we obtain $$\frac{\lambda_B\sqrt{V_g}}{10^3 r_g}=4{\cdot}45\sqrt{\frac{\{r_u^2/r_g^2-1\}}{(i_g/i_0)\,G_u}} \qquad \text{.....(20).}$$

In practice it will be found that $\{r_u^2/r_g^2-1\}/G_u=2$ approximately for all values of i_g/i_0; whence

$$\frac{\lambda_B\sqrt{V_g}}{10^3 r_g}=\frac{6{\cdot}30}{\sqrt{i_g/i_0}}\text{ approximately} \qquad \text{.....(20}a\text{).}$$

When the grid-plate space becomes unsaturated r_u remains constant and equal to r_p, but the transit time, represented by G_u, will decrease. Thus as i_g/i_0 is reduced below the value 0·3 for valve 6, at which grid-plate saturation occurs, the wavelengths obtained in practice will be longer than given by equations (20) and (20*a*).

The theoretical curve calculated from equation (20*a*) is shown in figure 12, together with the experimental curve of valve 6.

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DISCUSSION

Dr D. Owen: The author has introduced order into the apparently complex data regarding electronic oscillations in a triode valve. These oscillations appear to be divisible into two fundamental types. When Barkhausen and Kurz announced their discovery of electronic oscillations, they proposed a transit hypothesis for the mechanism of the oscillations. Much later Rostagni suggested the electron-plasma or electron-cloud hypothesis. It appears that the author's A type of oscillations accords with Barkhausen's mechanism, and the B type with Rostagni's. The Gill-Morell oscillations appear to be oscillations of great amplitude which occur when the external circuit is in resonance with the A type. It is not very clear, however, why a similar enhancement of amplitude should not occur in connexion with the B type, or what is the real significance of the connexion (as shown in figure 11) between Gill-Morell oscillations and the oscillations of type B. It would be useful if the author could make these points clearer.

L. Hartshorn: It struck me as very curious that in order to account for oscillations of type A we are asked to think of the time of transit of electrons between two equipotentials, while to account for those of type B we are referred to an equivalent inductance of the electrons and the capacitance between two equi-

potentials. Why is one method of approach applicable to oscillations A and not to B, and vice versa? I cannot help feeling that both oscillations A and B should be explicable in terms of a mass factor (electron inertia) and a spring factor (distortion of the electric field produced by their motion), and that an arbitrary difference of mental picture should not be necessary in the approach to the two modes of oscillation. What is the difference between them? Possibly A corresponds to the oscillations of the individual electrons, while B corresponds to oscillations of the distribution pattern of the electron stream. Even when we have accounted for the existence of various natural frequencies of the valve in some such way, we still have to account for the maintenance of the oscillations, probably in terms of a so-called negative resistance.

E. C. S. Megaw: The outstanding results of the author's work seem to me to be (1) confirmation of the view that coherent oscillations can in some circumstances be maintained by an electron discharge without any electrical resonant circuit other than the electrons themselves; and (2) the indication that in a positive-grid triode there are at least two possible modes or degrees of freedom for such oscillations. As regards (1), although I disputed this point with the author some years ago, subsequent experiments seem to have established the correctness of his view beyond question. The mechanism of maintenance of coherent, or synchronized, oscillations of this kind is far from clear. A periodic variation of potential somewhere in the system would seem to be essential, and if the impedance of the external circuit is negligible in comparison with that of the discharge path, it is difficult to imagine how such a variation could arise unless space-charge limitation occurs in either the filament-grid or the grid-anode space. The author's results are apparently consistent with the supposition that these oscillations are dependent on the existence of space-charge limitation. The mechanism of these oscillations may well be similar to that of Tonks and Langmuir's plasma-electron oscillations.

As to point (2), while the author has certainly shown that there are two possible modes of oscillation, his argument to the effect that there are two different mechanisms seems less secure; and the extrapolation carried out in figure 11 is based on what may, after all, be only a coincidence. At all events it does not convince me of the validity of Rostagni's theory, which I believe is fundamentally in error in applying equations derived for statistical equilibrium to current-flow in the grid-anode space. It can also be shown by direct measurement that the effect of electron-inertia in this case is not represented by the addition of a pure inductance to the circuit.

Author's reply: With regard to the points raised by Dr Owen, it is necessary to consider the conditions for maintenance of electron oscillations. It has been shown theoretically by Mr Benham that a stream of electrons passing between two electrodes exhibits the property of negative resistance for certain narrow ranges of frequency, with a maximum value for a definite wave-length in each range. For the Gill-Morell oscillations maxima in amplitude will be obtained when their wave-length corresponds with maximum negative resistance: but maxima

are also to be expected when the external circuit resonates with the equivalent circuits of the type-A and type-B oscillations to which it is coupled. The optima considered in figure 11 are due to resonance with the type-B circuit. Then enhancement of amplitude of the Gill-Morell oscillations, shown in figure 2, which appears to occur for resonance with the type-A circuit, is most probably due to correspondence with the maximum negative resistance.

In reply to Dr Hartshorn: The experimental evidence necessitating the different explanations for the two types of Barkhausen-Kurz oscillations is summarized in § 6. Provided that the space between two electrodes remains saturated, the electron-density varies inversely as the square of the transit time, and the two explanations might then be regarded as equivalent, at least qualitatively; but when, with reduction of emission current, the space becomes unsaturated, the relation no longer holds. The transit time decreases only slightly, whereas the electron-density decreases to zero, and it is then possible to distinguish between the two explanations.

The reason for the existence of wave-lengths determined by such different mechanisms appears to be closely linked with the conditions of maintenance, and I hope to deal with the question of oscillation-maintenance more fully in a further paper. While I agree with Mr Megaw that space charge appears to be the ultimate cause of maintenance, I do not consider that space-charge limitation of current is essential. The results shown in figure 8, and the fact that the range of values of i_g/i_0 for which Barkhausen-Kurz oscillations are obtained does not appear to depend on plate-radius, support this view.

As regards Mr Megaw's second point, I must admit that I am not entirely satisfied with the details of Rostagni's theory, but I am of the opinion that for oscillations of small amplitude the assumption of statistical equilibrium is sufficiently close to the truth, and gives an explanation which is at any rate qualitatively correct.

AN X-RAY GONIOMETER USING BEAMS OF LARGE APERTURE FOR PHOTOGRAPHICALLY RECORDING CRYSTAL-POWDER REFLECTIONS

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ABSTRACT. The factors which determine the various aberrations of X-ray reflections from microcrystalline powders are considered in relation to the design of a goniometer using beams of large aperture. Expressions are derived for the aberrations from a flat layer and from a toroidal powder surface in relation to the angular setting. An instrument using wide beams is described in detail and indications are given regarding its operation. According to the procedure adopted such aberrations as lead to asymmetrical diffusion effects are reduced, and in this way sharp lines with correctly located peaks are obtained. Powder layers with spherical and toroidal surfaces are discussed in detail and data are given for their use; as compared with flat layers they make it possible to employ considerably wider beams with correspondingly shorter exposures, particularly in the range of medium and large angles.

§ 1. INTRODUCTION

The essential requirement which any X-ray goniometer must satisfy is that it must give distinct separation of X-ray beams according to the angles through which they are deflected by a crystal lattice. Any such angle is determined by the relation

$$\sin\theta=\frac{n\lambda}{2d} \qquad \ldots\ldots(1),$$

where θ is the glancing angle of incidence and of reflection with respect to the lattice planes, d their spacing, λ the wave-length and n an integer. For a given wave-length different angles of deflection 2θ are then associated with the different spacings. For any spacing or reflection which for a given wave-length is represented by a definite value of θ, a parallel monochromatic pencil will be deflected from a microcrystalline powder along the surface of a cone of aperture 4θ.

In a powder camera of the Debye type generally used a narrow X-ray pencil falls on a powder rod and the reflections are recorded as lines with an angular width of the same order as the width of the incident beam. Good definition of the reflections can then be obtained only by the use of very narrow beams.

Attention has been drawn by the writer[1, 2] to the possibility of using beams of greater angular width with correspondingly shorter exposures, by disposing the powder on such a surface that all elements of it can contribute to deflect radiation through a fixed angle 2θ to one point.

A method developed from these considerations has been used in a number of determinations and was shown to present certain advantages not only in reducing the times of exposure required for the evaluation of angles, but also in establishing particularly simple conditions for quantitative measurements. In these *Proceedings*[3] an account has been given of the technique developed for the measurement of intensities of reflection; attention was then drawn to the importance of being able to satisfy the conditions for good angular definition in quantitative measurements, a point which will be discussed further in a forthcoming publication by Baxter and the writer. The method for obtaining well-defined deflections as required *inter alia* for accurate angular measurements and the instrument in its present form have not yet been described. This method consists essentially in distinguishing between various aberrations according to whether the diffusion they produce is symmetrically or asymmetrically distributed and using the instrument so as to reduce asymmetric effects to a negligible amount. In this way reflection lines with remarkably correct peaks are obtained.

In order to give an account of the procedure adopted we first discuss the various aberrations or diffusion effects encountered in crystal reflections from powders, with a view to collecting those expressions which are used in the operation of the instrument.

§ 2. THE ABERRATIONS ASSOCIATED WITH X-RAY REFLECTIONS FROM POWDER SURFACES

The surface on which we consider the powder particles to be placed is not a reflecting surface in the sense of an optical mirror, i.e. it does in general not represent the orientation of the lattice planes at which reflection takes place, but has the

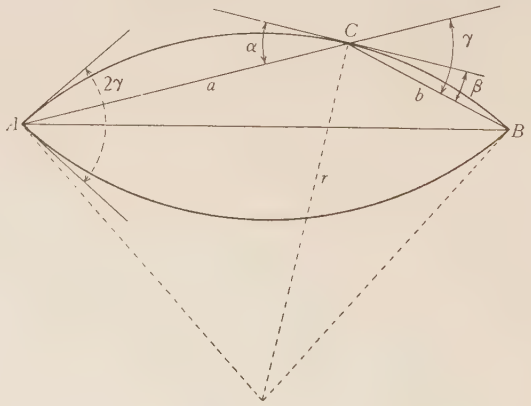


Figure 1.

property, which meets the particular case, that lines drawn from two points A and B to any point C on it, figure 1, make an angle in C corresponding to a constant deflection at the surface. It is obtained by rotating an arc with the peripheral angle $2(\pi-\theta)$ about its chord; the points A and B are then the intersections of the arc with the axis of rotation. This surface is here called a *toroidal surface.*

This toroidal surface constitutes the geometrical locus of the points from which X rays emerging from A are deflected to B, through an angle of deflection $\gamma = 2\theta$. The aperture of the beam which can be so deflected is determined by a cone of aperture $2\gamma = 4\theta$, and this represents the maximum angular aperture through which radiation emerging from one point can be deflected by any arrangement to another point with a fixed angle of deflection 2θ.

From equation (1) a different θ and a different toroid is associated with each value λ/d, so that with a given spacing d any particular toroid will act as a monochromator for one wave-length only. A pattern corresponding to the various reflections of one wave-length according to the different spacings of the crystal particles which is of more general interest cannot be obtained by a single extended surface of this type. It is however useful to refer to the toroidal surface in comparing the efficiency of various arrangements and in determining their limits of definition.

An annular zone of the toroid and a zonal band have been used by the writer[4] and by Bohlin[5] respectively. Applied to photographic recording, these arrangements are unfavourable because the scattered X rays do not fall normally on the photographic film and the reflections are produced by pencils involving different magnifications between source and image.

In the present instrument which corresponds to an arrangement outlined in a previous paper[2] an X-ray beam diverging from A falls on a powder area in C, figure 2, which can be rotated about a vertical axis; the scattered radiation is recorded on a cylindrical film disposed on a circle with C as centre. In this way we obtain normal incidence of the reflected X rays upon the photographic film and constant magnification between the source and its reflections on the film; and at the same time we can approximate to the toroid corresponding to any value of θ by setting the centre of the powder surface in C tangential to it; this is satisfied when

$$\left.\begin{aligned} \frac{\sin\alpha}{\sin\beta} &= \frac{a}{b} \\ \text{with} \quad \alpha+\beta &= 2\theta \end{aligned}\right\} \qquad \ldots\ldots(2),$$

where α and β are the glancing angles of the incident and reflected beam and a and b the distances of the source and film from the layer. For an extended range of θ's corresponding to a certain range ζ on the film the powder is rotated and a diaphragm D, figure 2, moves with the powder surface, so as to uncover in succession only a small area of the film for which equation (2) is satisfied.

The sharpness of the reflections depends then on certain geometrical conditions. (i) With a powder layer correctly set for a reflection angle θ_1, and a point B_1 on the film, any other reflection with an angle θ_2 corresponding to a point B_2 will show a certain diffusion, which increases with the distance of B_2 from B_1 since the setting of the powder layer corresponds to the orientation of the toroid associated with θ_1 and B_1 and not to the one association with θ_2 and B_2. With a rotating diaphragm this diffusion is produced to the extent to which the layer is permitted to rotate outside its correct setting before the radiation is intercepted by the diaphragm. (ii) With a point source at A and the centre of the powder layer tangential to the toroid corre-

sponding to a certain angle θ_1, and to the point B_1, the elements of the layer outside the centre will reflect the radiation deflected through $2\theta_1$, to points slightly away from B_1 in so far as the particular elements of the layer do not coincide with the toroid, but are at a small distance from it. (iii) For points on the film above or below the plane ACB radiation deflected from the centre of the layer through any particular fixed angle 2θ will trace a line corresponding to the intersection of a cone of aperture

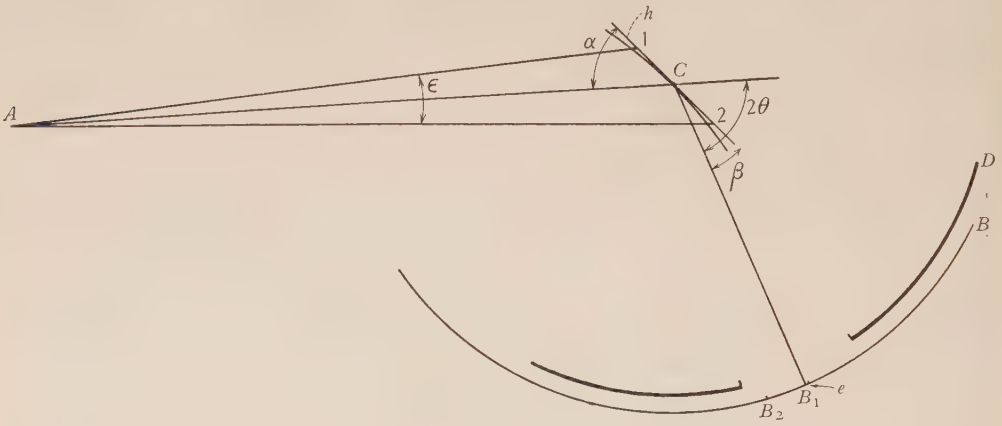


Figure 2.

4θ with the film. For values of 2θ either small or near 2π these lines approximate to circles, which show in general increasing diffusion for points farther away from the plane ACB, owing to the greater discrepancy between the orientation of the powder surface and the orientation of the toroids associated with the particular points of the film outside its centre line. (iv) If the point source in A is replaced by a slit normal to the plane ACB, the reflection with which 2θ is associated is scattered from the central part of the layer not as one cone, but as a range of cones with axes corresponding to the directions of the incident beam. Instead of one ring we have thus a system of rings, figure 3, with centres displaced along a line EF normal to plane ACB. This

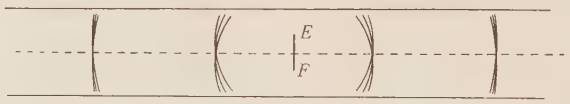


Figure 3.

produces a certain diffusion and introduces a serious error into the quantitative evaluation of intensities at small angles and at angles approximating to 2π, since the microphotometric evaluation of a line involves an error depending on the extent to which the line is crossed obliquely. This effect and the effect under (iii) above apply to all powder arrangements and are not peculiar to our method. They are well known and can easily be calculated. We take account of them by avoiding elongated slit systems and by reducing the angular height of the beam for small angles. They do therefore not need to be considered here further.

§ 3. EXPRESSIONS FOR THE ABERRATION DUE TO ANGULAR DISPLACEMENT OF THE TOROIDAL SURFACE

The magnitude of the effects discussed under (i) and (ii) in § 2 depends on the surface occupied by the powder, on the angles, and on the aperture of the incident beam.

The linear displacement e of the reflection on the film, which occurs when the reflecting point is situated outside of the toroidal surface of correct definition at a distance h, is given by the projection of h on the surface of the photographic film (see the case illustrated by figure 2) and thus by

$$e = h \sin 2\theta / \sin \alpha \qquad \text{.....(3),}$$

or in angular measure

$$\omega = h \sin 2\theta / b \sin \alpha \qquad \text{.....(4).}$$

Applied to the diffusion effect discussed in § 2 under (i), owing to the scattering surface being turned by an angle δ out of the position for which it represents the correct toroid, we find that a scattering point at the distance l from the axis of rotation is displaced by $h = \delta l$ and considering an incident beam of semiaperture $\epsilon/2$ the greatest distance becomes

$$l = \tfrac{1}{2}\epsilon a / \sin \alpha$$

and the greatest displacement

$$h = \tfrac{1}{2}\delta\epsilon a / \sin \alpha ;$$

on introducing this into equation (3) and multiplying by a factor 2 to obtain the added effect of the two sides (1 and 2 in figure 2) of the layer, we obtain

$$e_1 = \delta\epsilon a \frac{\sin 2\theta}{\sin^2 \alpha}$$

and, to the degree of accuracy with which we can substitute for a from equation (2),

$$e_1 = \delta\epsilon b \frac{\sin 2\theta}{\sin \alpha \sin \beta} \qquad \text{.....(5),}$$

or in angular measure

$$\omega_1 = \delta\epsilon \frac{\sin 2\theta}{\sin \alpha \sin \beta} \qquad \text{.....(6),}$$

where α and β represent the correct angles satisfying equation (2). Equations (5) and (6) measure the extreme limits of diffusion associated with an angular displacement δ of the layer out of the orientation of correct focusing about an axis normal to ACB. We see that for a given reflection 2θ the latitude allowed in the setting of the powder so as to comply with a certain permissible diffusion is greatest when $\alpha = \beta$ or $a = b$, but varies little with the ratio $a : b$ as long as this is not far from unity. These relations also show how the latitude in setting varies with θ. For $a = b$ we have from equations (2) and (6)

$$\omega_1 = 2\delta\epsilon \cot \beta = 2\delta\epsilon \cot \theta \qquad \text{.....(7),}$$

which indicates that the latitude in setting increases with increasing angle as $\cot \theta$ becomes smaller.

If instead of an angular displacement δ of the layer we consider a corresponding displacement ζ on the film, we obtain the diffusion for a point distant by the angle ζ from the point for which the layer is correctly set. Corresponding rotations of the powder and displacements of the point of correct definition on the film are interlinked by equation (2); from this by differentiation, since for any angle of deflection $\gamma=\alpha+\beta$, we have

$$\zeta=\left(1+\frac{b\cos\alpha}{a\cos\beta}\right)\delta,$$

and using equation (2)

$$\zeta=(1+\cot\alpha\tan\beta)\,\delta;$$

and thus for the angular width of the diffusion

$$\omega_1=\epsilon\zeta\,\frac{\tan\alpha}{\tan\alpha+\tan\beta}\,\frac{\sin 2\theta}{\sin\alpha\sin\beta} \qquad \text{.....(8).}$$

For small angles, when $\cos\alpha$ and $\cos\beta$ are both nearly unity, and the sines can be substituted for the tangents, we can write

$$\zeta=\frac{a+b}{a}\,\delta$$

and thus

$$\omega_1=\epsilon\zeta\,\frac{a}{a+b}\,\frac{\sin 2\theta}{\sin\alpha\sin\beta} \qquad \text{.....(9),}$$

while for $a=b$

$$\omega_1=\epsilon\zeta\cot\theta \qquad \text{.....(10).}$$

If a rotating diaphragm be used and moved so that the centre of the opening of aperture η satisfies equation (2) for all angles, then the extreme displacement of the powder out of its correct setting before the radiation is intercepted by the screen corresponds to $\zeta=\eta/2$, the limit of diffusion is thus

$$\omega_1=\tfrac{1}{2}\epsilon\eta\,\frac{1}{1+\cot\alpha\tan\beta}\,\frac{\sin 2\theta}{\sin\alpha\sin\beta} \qquad \text{.....(11),}$$

which for $a=b$ becomes

$$\omega_1=\tfrac{1}{2}\epsilon\eta\cot\beta=\tfrac{1}{2}\epsilon\eta\cot\theta \qquad \text{.....(12).}$$

§ 4. EXPRESSIONS FOR THE ABERRATION DUE TO THE REFLECTING SURFACE NOT CONFORMING WITH THE TOROID

The diffusion effect considered in § 2 (ii) and due to the scattering surface not conforming with the toroid can be rendered by simple expressions for the special case of a flat surface, which is of particular interest with regard to 'powders' consisting of a coherent microcrystalline aggregate, on which by grinding and cutting flat surfaces are most easily obtained. For a point of the flat surface in the plane ACB the distance h from the toroidal surface is

$$h=l^2/2r \qquad \text{.....(13),}$$

where l is the distance of the point from C and r is the radius of curvature of the toroid in the plane ACB. Introducing the value of $r=a/2\sin\alpha$, see figure 1, in equation (3), and remembering that $l=\tfrac{1}{2}\epsilon a/\sin\alpha$, we have

$$h=\left(\frac{\epsilon}{2}\right)^2\frac{a}{\sin\alpha}$$

and for the angular diffusion

$$\omega_2 = \left(\frac{\epsilon}{2}\right)^2 \frac{a \sin 2\theta}{b \sin^2 \alpha},$$

and when equation (2) is satisfied

$$\omega_2 = \left(\frac{\epsilon}{2}\right)^2 \frac{\sin 2\theta}{\sin \alpha \sin \beta} \qquad \ldots\ldots(14),$$

which measures the extreme diffusion to one side of the point of correct definition. Similarly with regard to the divergence ϵ' in a plane normal to ACB we consider that for points of the layer above or below plane ACB the radius of curvature of the toroid in a plane normal to ACB is given by $r' = a \sin \alpha = b \sin \beta$ (approximately when a is not very different from b). Substituting this in the equation corresponding to (13) and in equation (4), we have

$$\omega_2' = \tfrac{1}{2}\left(\frac{\epsilon'}{2}\right)^2 \frac{\sin 2\theta}{\sin \alpha \sin \beta} \qquad \ldots\ldots(15).$$

In these expressions $\epsilon/2$ and $\epsilon'/2$ are assumed to be small compared with α. More general expressions covering the case of α and ϵ respectively, ϵ' being of the same magnitude, are without interest, because for the reasons discussed in § 2 (iv) only narrow beams can be used for small angles. For small angles the beams incident at points nearer to and farther from the source (points 1 and 2 in figure 2) give different diffusions. Asymmetric beams can then be used to give the optimum condition so that the general discussion based on beams symmetrical with respect to the axis AC is of limited interest.*

The diffusion discussed in § 2 (i) and in § 3 and associated with an angular displacement of the powder out of its correct orientation, is expressed by essentially the same relations whether the surface is the actual toroid or a surface approximating to it; in the latter case other diffusion effects and in particular the diffusion considered in § 2 (ii) and in the present paragraph are superposed. There is a considerable advantage in reducing the latter, which is due to lack of conformity between the powder surface and the toroid, since it is in general very unequally distributed with respect to the true position of the line. In the case of the flat layer, for instance, it is all extended towards one side except in extreme cases, when a and b are unequal. This diffusion therefore affects the location of the lines very adversely, whereas the diffusion discussed in § 2 (i), due to the angular displacement of the layer out of its correct setting, is found to be very nearly symmetrically distributed for practically the whole of the angular range when the incident beam conforms with the conditions here assumed.

With curved powder layers the toroidal surface can be approximated to very closely and the asymmetric diffusion discussed in § 2 (ii) can be reduced. The rotational displacement is then the main aberration to be considered.

From the expressions (5) to (12) where ϵ appears to the first power we see that

* In the earlier brief discussion[2] of these relations we considered a and b to be approximately equal; for $a=b$ and $\alpha=\beta$ when $\sin 2\theta/\sin \alpha \sin \beta = 2 \cot \beta$ we obtain $\omega_2=(\epsilon^2/2)\cot\beta$, while in the earlier paper we gave $\omega_2=(\epsilon^2/4)\cot\beta$, a factor 2 having erroneously been omitted. Similarly from expression $d=b\,\delta\epsilon\cot\beta$ of the early paper we derived $\omega_1=\delta\epsilon\cot\beta$, which measures the width of the diffusion from the centre to one edge, while the total width according to our present expression (7) is $\omega_1=2\delta\epsilon\cot\beta$.

with circular apertures of the incident beam the falling off in intensity from the centre of the line is slow at first and then more rapid. In conjunction with the width of the slit in A actually a more uniform decrease of intensity results. When the slit-width is approximately equal to the linear diffusion as expressed by equation (5), lines with a well-marked narrow peak, which are most suitable for determining angles, are obtained. When $a=b$ a concave spherical surface with radius $b/\sqrt{2}$ can be used as a first general approximation to the toroid. Much better approximations are obtained over definite angular ranges with surfaces of double curvature.

The expressions (6) to (12) and (14), (15) can be used to give directly the numerical values of the diffusion in radians if the quantities ϵ, δ, ζ and η entering into them are expressed in radians. If these are given in degrees they have to be divided by the unit of angular measure, namely 57·3°. The expression (5) for the linear width gives this in the units in which b is measured, when numerical values of δ and ϵ in radians are again introduced.

§ 5. THE MAGNIFICATION EFFECT

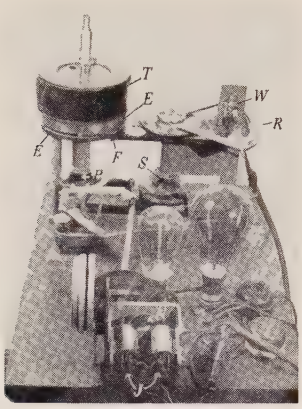
The intensity of reflections has been discussed in another paper[3]. *Ceteris paribus* it depends on the volume of the powder from which X rays are scattered, and this leads to a term $1/\mu$ as regards the absorption coefficient μ and for a flat or quasi-flat layer to a term

$$i=\sin\beta/\cos\tfrac{1}{2}(\beta-\alpha) \qquad \text{.....(16),}$$

as regards its orientation. The intensity depends then further on the angle of reflection and on the scattering properties of the particular crystallites. These relations have been discussed elsewhere. Mention should be made, however, of a geometrical-optical magnification effect between the collimator slit or source and the reflections on the film, which is of particular significance in the operation of the present method. As long as the aberrations are sufficiently small the width of the reflection lines is controlled by this magnification factor and varies with the ratio b/a with a gain of specific intensity when the ratio is decreased. This gain is partly compensated by the decrease of i, equation (16), owing to the decrease of β which follows an increase of a/b for any fixed angle 2θ. We have thus from these relations

$$\epsilon\epsilon' i a/b^2 \qquad \text{.....(17),}$$

as an intensity expression indicating how the specific intensity of the reflections, which determines the photographic density of the lines, depends on the quantities entering in the setting of the instrument. Term $\epsilon\epsilon'$ assumes an elliptical incident beam, for which the solid angle subtended is $\pi\epsilon\epsilon'$. Term a/b^2 is adapted to approximate to the particular conditions of recording the reflections, where the lines are projected on a circle of radius $b\sin 2\theta$ which introduces a factor $1/b$, while the geometrical-optical magnification effect owing to superpositions introduces a factor a/b and not $(a/b)^2$. With an incident X-ray beam of intensity X per unit solid angle and a slit in A of cross-section σ, the specific intensity of the reflections will be proportional to X or to $X\sqrt{\sigma}$ according to whether the width of the lines can be considered as proportional to the area σ of the slit or, in the case of geometrically



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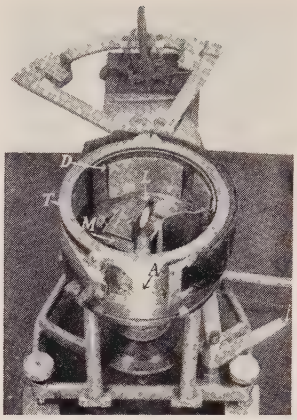

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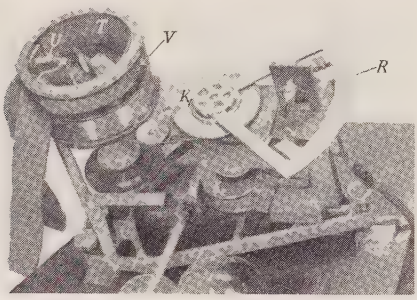

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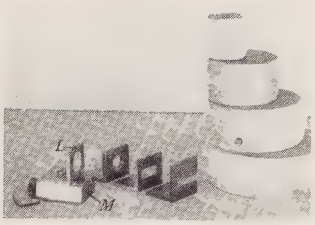
Figure 4.



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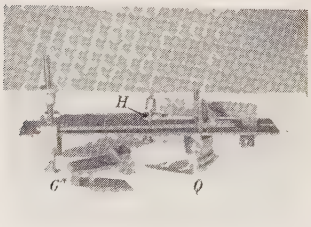

b

Figure 5.

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similar slits, to one of its linear dimensions and thus to $\sqrt{\sigma}$ only. When, however, the width of the lines is essentially determined by the limited resolving-power of the crystallites or by the aberrations and not by the entrance slit, then the specific intensity is proportional to $X\sigma$. The conditions will be chosen so as to meet each particular case. For routine crystallographic work it is convenient to arrange for the aperture of the entrance slit and the aberrations both to contribute to the width of the lines. When on the other hand the size distribution of the crystallites constituting a powder is under investigation, both the aperture of the slit and the aberrations are reduced to such an extent that the resolving-power of the crystallites becomes the essential factor in determining the width of the lines.

§ 6. DESCRIPTION OF THE GONIOMETER

The instrument comprises essentially two parallel shafts mounted in ball bearings to a platform. One of these shafts P supports on its upper end the holder for the powder layer L. Concentric with it are the rotating diaphragm D and the camera ring T, which supports the film and the collimating slit system. Diaphragm D is supported on an annular turntable, also mounted in ball bearings. The camera ring fits on a support E on which it is definitely located. The support E can be rotated and clamped in any position by clamping screws F. The axis of rotation of the camera ring with its support coincides very exactly with the axis of the shaft P. In the actual making of these parts, shaft P was used as a mandril to ensure that the various rotating parts should be truly concentric. Flat pulleys of equal diameter are fixed to the lower part of P and to the support of diaphragm D, and steel wires run from these to the driving shaft S. The latter carries at its upper part a pulley K_1 corresponding to the pulley at the rotating diaphragm and of the same diameter, so that both move at the same angular speed. The driving shaft carries a pulley or a cam K_2 corresponding to the lower pulley fixed to shaft P. When the distances a from slit to powder and b from powder to film are equal, so that the angular velocity of the powder layer should be half that of the diaphragm, the pulley K_2 on S has half the diameter of the corresponding pulley on P. For different ratios $a : b$ this pulley is replaced by a cam, so as to satisfy the correct rate of rotation of powder and diaphragm in accordance with relation (2). In many cases a sufficient approximation is obtained from a circular pulley excentrically mounted on S. The steel wires are held in tension by spring loaded jockey pulleys as shown in figure 4*c*.

The motion of the driving shaft S is obtained through a worm drive engaging in a wheel at its lower end, and the range of rotation and reversal of motion is controlled by a large sector R clamped at the upper end. Two movable cams on this sector, which can be adjusted to the desired range of rocking, operate a mercury switch W and reverse the direction of rotation of the worm. Figure 4*a* indicates the arrangement of the gears. The reversal is produced by the shift of a dog clutch operated by electromagnets which engages with one or the other of two cog wheels placed on opposite sides of the crownwheel N. The mercury switch controls the electromagnets J which are in shunt with a carbon filament lamp and a metal filament lamp is placed in series. This combination provides a large current and

strong pull by the reversing magnets for the reversal after the current has been momentarily broken by the swinging over of the switch and afterwards provides a weak current, sufficient to keep the wheels engaged. This system of drive with steel wires was not only found convenient for readily obtaining the varying ratios of motion demanded for different values of a and b, but in conjunction with ball bearings for all moving parts was found to give steady motion which is most essential for quantitative measurements with a minimum of back-lash. The mechanically operated switch with an electromagnetic gear-change was found reliable and never gave a failure during two years of use.

The camera ring T is turned from a heavy casting. The film is pressed by a soft packing against a cylindrical surface of radius 5·73 cm., so that 1 mm. on the film surface corresponds to 1°. It is covered on the outside with lead rubber. The X rays reach the film through slots with characteristic fiducial marks at intervals of 12°. The collimating and slit system consists of tube U, which screws into the camera ring and is kept central very exactly by a large flange. Lead diaphragms mounted in brass, which define the aperture of the beam, can be inserted in both ends of this tube. For $a=b$ the entrance slit in A consists of two lead jaws mounted in brass pieces, which fit into the circular recess receiving the film. The height of the slit is determined by a circular diaphragm inserted in U, immediately behind the slit, while a larger circular or elliptical diaphragm at the end near the powder defines the divergence of the rays. When $a>b$, tube U can be reversed and fitted on a bridge piece, not shown in the figures, on the outside of the camera ring so that the film in its recess can be inserted all round it. In this position tube U supports the entrance slit at its outer end, while the end turned towards the camera ring receives the wider diaphragm. The interior of the camera is then entirely free from any obstruction. Opposite tube U a circular aperture V is made in the camera ring and is plugged with a celluloid disk holding a small lead ball at its centre, which serves for accurate collimating.

The fixing of the collimating diaphragms to the camera ring in conjunction with the definite location of the powder layer ensures in the simplest way that in a series of exposures taken under varying conditions, the same lines shall always take the same position with respect to the fiducial marks even when the various parts are reset. In fact it is possible to interrupt an exposure and to continue it after resetting the powder or the camera ring and collimator. The fixing of the camera ring on a support, which can be rotated, facilitates the setting of the instrument with regard to the X-ray source. This is done by first removing the camera ring and setting the instrument so that the centre of the X-ray beam falls on the position taken by the powder layer. Then the camera ring is inserted and its support unclamped. It can now be rotated until the beam passing through the collimating tube U appears brightest. Exact setting is obtained by observing on a fluorescent screen the beam at the back of the camera with the powder layer removed. The setting is correct when, with a small circular diaphragm in U, the lead ball appears as a dark centre surrounded by a narrow fluorescent ring. This setting is obtained by means of two independent operations: rotation of the camera ring about a vertical axis and

tilting of the camera by the motion of the back set-screw. This adjustment is quicker and utilizes the best part of the beam with greater certainty than does the positioning of a camera with a slit system fixed to the instrument, when lateral displacements and rotations of the instrument have to be combined.

The rotating diaphragm D, figure 4*b*, consists essentially of a shield with an opening for the passage of X rays mounted on a ring with a graduated scale, which fits on the rotating platform. The opening of the diaphragm can be adjusted to the desired value of η. On the side near the collimating tube a flexible extension is provided allowing the diaphragm to be used for large angles of deflection and providing shielding for the film, when the diaphragm is moved away to smaller angles.

The support of the powder layer consists of two parts M and L. The clamping block M in figure 5*a* fits on shaft P and carries a graduated scale. Its top surface is at right angles to the axis of P and the powder holder is correctly located on M by means of two screws, which mark the line passing through the axis of rotation. The holder L consists essentially of a piece of metal of ⌊ form, the outside surfaces of which are accurately machined at right angles, and with an opening on one side for the powder. A number of these holders of simple form with openings to fit the various surfaces are shown in figure 5*a* together with surfaces of double curvature used as moulds. To prepare the powder layer the holder is placed with its opening against a mould of the desired curvature and the powder, mixed with some binding material, is introduced in a crumbly or pasty state from the back. It is essential, at least for quantitative work, not to scrape or polish the front surface of the powder as this produces preferential orientation similar to that found with layers prepared from the front side, which are useless for quantitative measurements. In the case of concave surfaces the lower edge which fits against the locating screws is recessed, so as to bring the centre of the layer over the axis of rotation.

Graduated scales on the support of the powder layer and on the rotating diaphragm are used to establish the correct angular setting with respect to the collimating tube. They are read off against reference marks on the camera ring. In the particular instrument illustrated, this is done by means of a small telescope with cross wires, fixed on the lid of the camera, which can be set in definite positions with respect to the collimating tube U. It can be made to view both scales, which are nearly at the same level and are illuminated by a small internal electric lamp. In this way the setting of the powder and of the diaphragm and their relative motion can easily be controlled.

Single crystals are set on the support M, optically adjusted and then inserted on shaft P for the exposure.

§ 7. THE MEASUREMENT OF ANGLES OF DEFLECTION

When a group of lines comprised within a comparatively narrow angular range is to be recorded no rotating diaphragm need be used. The powder layer is set so as to conform with equation (2) for some intermediate angle and the aperture of the beam then adjusted to give the required definition over the extent of ζ on either side of the focused angle determined by equations (8), (9) and (10). Owing to the diffusion

effect considered in § 2 (i) the lines are then not equally sharp, those near the angle for which the layer is correctly set being sharper and narrower. When reflections of unequal intensities are being recorded it is often possible to take advantage of this by choosing the angular setting so as to make the weak lines narrower and thus enhance their specific intensities and their peak values, as was discussed in relation to quantitative intensity-measurements in a previous paper[3]. With a flat layer we have also to consider the limits of aperture imposed by the effect of § 2 (ii), equations (14) and (15). With a curved layer it is generally possible to approximate to the toroidal surface, for the particular angular range, so closely that this effect does not need to be considered in determining the aperture of the beam. Table 1 gives values for the principal radii of curvature of the toroidal surface for $a=b=5{\cdot}73$ cm. and for $a:b=1{\cdot}5$ with $a=8{\cdot}60$ cm. and $b=5{\cdot}73$ cm. for various values of 2θ; r is the radius of curvature in the plane ACB, and r' that in the plane normal to it.

Table 1

2θ (degrees)	$a:b=1$		$a:b=1{\cdot}5$	
	r (cm.)	r' (cm.)	r (cm.)	r' (cm.)
20	16·3	1·0	20·6	3·5
30	11·0	1·5	13·8	3·4
45	7·5	2·2	9·8	3·4
60	5·7	3·0	7·2	3·5
90	4·0	4·0	5·1	5·1
120	3·3	5·0	4·4	7·1
150	3·0	5·5	4·6	18·7

For practical purposes it is found that with the limits of aperture required to give good definition within a certain angular range having regard to other aberrations, the set of surfaces indicated by the table is adequate and in fact allows a sufficient overlap to meet the requirements of any angle of deflection without imposing any additional restriction on the permissible width of the beam. For very small and very large values of 2θ one or the other radius of curvature changes rapidly; this will then narrow down the angular range in which any particular surface can be used when the radius of curvature is small. This applies to r' with a equal to b in the region of small angles, but in this case the permissible vertical aperture ϵ' of the beam is reduced by the conical divergence effects considered in § 2 (iii) and (iv), so that any departure of the surface from the correct shape is less important. In exposures of this type a rocking motion is given to the powder layer through a range of about half a degree, in order to avoid spotted lines and to get better averages. An additional pulley is then inserted between shaft S and shaft P to reduce the angular velocity.

When it is desired to record reflections over a large angular range it is preferable to use the rotating diaphragm, with which all the lines are recorded under similar conditions. The apertures of the beam and of the diaphragm are then determined by the expression (11) or (12). A spherical surface will meet the more general case better than a flat layer, while for particular sections a surface of double curvature

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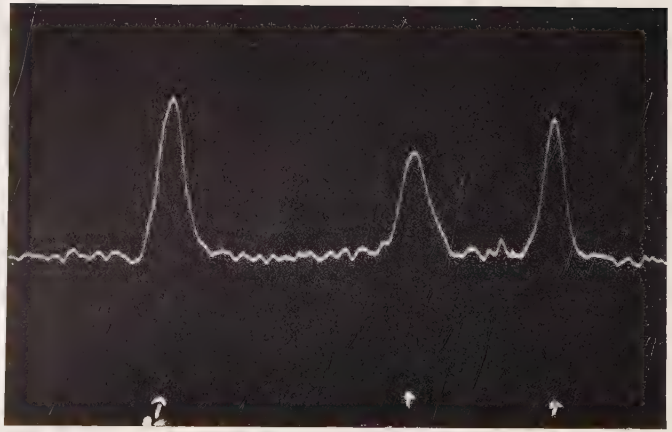

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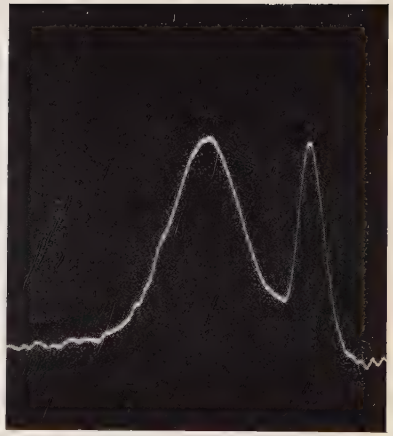

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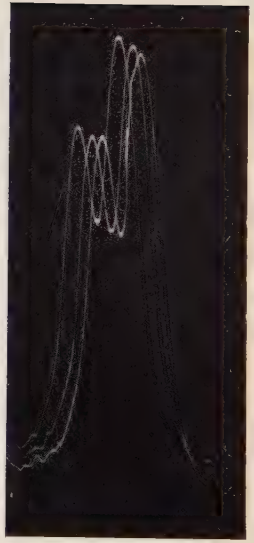

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e

f

Figure 6. (*a–d*) microphotometric records of lines. (*a*) (220) reflection from aluminium, set at focusing angle; $\epsilon = \epsilon' = 1{\cdot}5°$. Photometer magnification 26 times. (*b*) reflections from aluminium and cadmium oxide recorded with rotating diaphragm. The cadmium-oxide line (middle), from a preparation generally considered to give sharp lines, is distinctly broadened. Record taken with flat layer. Indication of asymmetry. (*c*) lines of cadmium oxide and platinum (broader line). The platinum powder is constituted of crystallites of varying size, so that the base of the line is broadened. A case of vanishing size-distribution. (*d*) K_α doublet of tungsten: fine powder with crystallites giving limited resolving-power, but little vanishing-size effect. Three records, slightly displaced, taken at different points of the line with very fine microphotometer slit, to bring out the correct shape of the line. (*e*) photograph of aluminium lines obtained from a spherical layer with a beam of large aperture; $\epsilon = \epsilon' = 10°$. (*f*) comparative patterns of aluminium and cadmium oxide lines photographed separately on the same film. The photometric records ($a-d$) were taken with a valve-bridge microphotometer, *Z. phys.* **70**, 74 (1931); *Phil. Mag.* **17**, 370 (1934).

can be used. Owing to the decrease of expressions (11) and (12) with larger angles of deflection it is preferable to approximate to the toroidal surface corresponding to an angle nearer to the smaller limit of the range used. The total time of exposure then increases with the range ν covered by the record, and can be given as $t\,(\nu/\eta+2)$, where t is the exposure received by any individual section. The additive term 2 comes from the side portions, which receive only a reduced exposure and are not used for quantitative measurements. For measurements of angles part of this side range can be utilized, so that the exposure is rendered rather by $t\,(\nu/\eta+1)$. The additive terms 2 and 1 thus involve an increase of exposure, which is particularly felt for small ν's, when it is useful to make η small as well and to combine it with a larger value of ϵ.

Considering more in detail the evaluation of angles, the method can be used to obtain sharp well-defined lines with comparatively short exposures. An example of such a line is given in figure 6*a*. A record covering an extended angular range obtained by the use of a rotating diaphragm and actually taken with an early instrument was reproduced in a previous paper[2]; we may refer to this reproduction to avoid repetition.

It should be noted that accurate determinations of spacings do not always require accurate angular measurements. Where it is practicable to use reflections for which, with the particular wave-length used, 2θ approaches 2π, then large changes of θ involve small changes of $\sin\theta$ and an approximate measure of θ is adequate. For this very reason, however, the angular range of θ to which this applies comprises only a very small latitude in the values of $\sin\theta$ and of the spacings d. It is for cases where a larger range of spacings has to be evaluated, which necessarily cannot all be comprised in the range of very large angles, that accurate angular measurements are required. Examples of the use of the method to meet such conditions are afforded by the determinations of the parameters of carbonates by Dawson and Adamson and by the writer[6,7]. Again, a high definition is essential when information as to the sizes of the crystallites constituting the powder particles is sought from the intensity-distribution of the individual lines, when it is essential to free the record of any other diffusion effect introduced by the goniometer. Figures 6*b* and 6*c* illustrate such examples.

The actual angles 2θ of the lines can be found in relation to the fiducial marks, subject to the satisfaction of definite geometrical relations between the slit system, the support of the film, and the powder surface. The present arrangement helps to secure these conditions and any deviations can be eliminated by calibration. The location of lines by reference to the edges of fiducial marks is however affected by the Ross effect and by other systematic errors and is not recommended when high accuracy is required; we prefer then to use a reference substance mixed with the powder and giving a pattern of lines for which the angles are well known as we have done in our measurements on carbonates[6]. This procedure also eliminates any line-displacement due to the penetration of the X rays into the layer.

§ 8. MEASUREMENTS WITH BEAMS OF LARGE ANGULAR APERTURE

In the commonest uses of the instrument an extreme increase in accuracy is not of as much interest as is a shortening of exposure, retaining a definition corresponding to the usual requirements of crystal-structure investigation. This can be satisfied with beams of considerable width, obtained by placing the point of divergence A of the rays on the anticathode itself. The distance a is then larger than b and this reduces the width of the image of the anticathode spot on the film and enhances the intensities in the manner discussed in § 5. The anticathode is set at a very small glancing angle and a screen with a cross slit attached to it cuts out radiation from points outside the anticathode spot. For such exposures the goniometer is firmly connected with the X-ray tube. Figure 6*e* shows a typical record taken with a spherical layer of aluminium when $a=8{\cdot}5$ cm. $b=5{\cdot}73$ cm. The recorded range extended from 60° to 110° while $\epsilon=\epsilon'=10°$ and $\eta=20°$. The total exposure was 1·5 mA.-hr., giving the actual exposure for any part of the film as 0·43 mA.-hr.

§ 9. THE QUANTITATIVE MEASUREMENT OF INTENSITIES

The technique of quantitative intensity-measurement by this method has been discussed in other papers[3, 8, 9]. It presents the advantage that by the use of powder surfaces of well-defined orientation with respect to the incident and reflected beams, conditions are established which lead to a simple quantitative evaluation of the observed reflections. With regard to the instrumental technique it is essential that the incident beam should be used at a constant aperture for the whole angular range exposed and further that for small angles of deflection the error due to the displaced ring patterns, to which we have referred in § 2 (iv), be avoided. These conditions have to be considered in choosing ϵ and ϵ'; generally ϵ' is then made smaller than ϵ. By satisfying closely the focusing condition and using a comparatively wide entrance slit in A, lines with a flattened peak and a sharp drop on both sides are obtained, and these are most suitable for quantitative measurements. When high accuracy is required conditions of extended diffusion due to defective focusing should be avoided, not only because diffused lines are unsuitable for accurate photometry, but also because the discontinuities of the background, to which attention has been drawn by Baxter and the writer, become correspondingly diffused so that overlapping results; accurate evaluation is then impossible.

§ 10. MODIFICATIONS OF THE INSTRUMENT FOR PARTICULAR REQUIREMENTS. REFLECTIONS FROM SINGLE CRYSTALS

In certain instances it is useful to increase the separation of the lines. This is done by increasing the distance b; the film is then held by an extension arm, which can be clamped to the camera ring. This is useful for small angles at which the intensity factor (16) increases with β and gives an advantage in satisfying equation

(2) when $b > a$. It partly compensates for the increase of exposure in projecting on a larger circle.

Figure 6 shows a device fitting in the camera ring by which reflections from two powder layers can be recorded on adjacent strips of a photographic film. Apart from applications in intensity-measurements to which we have referred in another paper[9], this device can be used in comparing corresponding lines of two powder specimens recorded under similar conditions. A grid Q can be displaced in front of the film by an amount equal to its spacing and according to its position uncovers alternate strips of the film. A swinging support holding two powder layers is displaced jointly with the grid by means of stem H, so that the reflections are recorded on separate strips. Figure 6*f* shows a record taken in this way. Brindley and Spiers[10] have described a similar device, in which however the reflections from two powder layers are superposed on the same strip.

The goniometer can be used to record the reflections from a single crystal and the rotating screen is then helpful in reducing the background by limiting the exposure of any particular part of the film to the time when the crystal is set so as to reflect radiation upon it. The conditions for reflection from the single crystal differ from those for a powder and the reflection from the particular set of lattice planes represented by the crystal face does not take place simultaneously from the whole of the surface, but for different points in succession as the crystal is turned. The width of the rotating diaphragm must be chosen accordingly.

§ 11. CAMERA DIMENSIONS AND ANGULAR DEFINITION

Apart from the geometrical-optical conditions here discussed, a certain diffusion is produced by the penetration of the X rays into the powder layer and by the scattering of the X rays in the photographic emulsion. These effects are well known and do not need further discussion. The former, which produces asymmetric lines, can be reduced where it is noticeable by adding to the powder an admixture of great absorbing power. Both these effects involve a certain linear broadening which affects angular measurements to a lesser extent when the camera-size is increased.

In this connexion it is of interest to consider the relation between the dimensions of the camera and the times of exposure. The angular accuracy depends upon factors controlling the angular diffusion of the beam, which for any given value 2θ do not contain the linear dimensions of the camera but only the apertures and the angles determining the setting of the powder. The intensity of the reflections is determined by the relations set out in § 5. Apart from the cases there considered we can say in a general way that with similar cameras, for a given total of scattered radiation, the specific intensity on the film is inversely proportional to the square of the dimensions. On the other hand, when a fixed value of definition is maintained, both linear dimensions of the entrance slit can be increased in proportion to the camera-size, which gives again a factor proportional to the square of the camera dimensions. We thus see that by complying with definite geometrical and optical relations, the times of exposure required for any given value of definition become

independent of the linear dimensions of the camera and no gain is obtained by making the camera small. This holds good as long as the full width of the entrance slit corresponding to a certain definition can be illuminated with X rays throughout the solid angle that can be utilized in the collimator. When the anticathode itself acts as slit, its width is fixed and then it is of advantage to reduce the size of the camera when high intensities with limited definition are required.

§ 12. RANGE OF APPLICATIONS OF THE METHOD

The method described in this paper has two distinct applications. One is the quantitative evaluation of the intensities of reflections and this has been discussed previously[3]. It had to be considered in the present account only in as far as the design and operation of the instrument must comply with the conditions set out in the more detailed discussion. The other application is the evaluation of the angles of reflection, and it has been shown that by a definite control of the aberrations as indicated in § 4, lines with correctly located peaks and a gradual falling off of intensity towards both sides are obtained, and these can be located with great accuracy. Where it is desired to push the accuracy very far, as in the instances referred to in § 7, incident beams with an aperture of 2° or 3° can be used; with curved powder layers an accuracy sufficient for the usual demands of structure-analysis can be attained in the medium range of deflections with beams of an aperture of 10° with correspondingly short exposures. In the region of very small angles, limits are set to the use of wide beams and this applies again to angles of deflection approximating to 2π, where the aberration considered in § 2 (iv) becomes important. For large angles the limitations introduced by this effect are not felt, since the evaluation of spacings is in that case but little affected by the limited accuracy of the angular measurement. Actually for angles of deflection approaching 180° the accuracy of the method becomes irrelevant for, in view of the smallness of the change in value of the sin function with angle for values near $\pi/2$, accurate determinations of spacings can be derived from comparatively coarse angular measurements. The particular field for the method in regard to determinations of spacings lies thus in the increase of accuracy with short exposures which can be attained in the medium angular range, extending from about 30° to 150°. The advantage of using this medium range of angles is that it accommodates a wide range of spacings; the ratio of the values of $\sin\theta$ associated with the angles of deflection 30° and 150° being 3·73, which implies that the largest spacing recorded in this range is over three and a half times larger than the smallest, while the range of spacings which can be recorded between 150° and 180° allows only a latitude of 3·5 per cent. It is for instances where lattice spacings of different length have to be evaluated and related, which cannot be accommodated on the narrow range offered at large angles, that the method has been developed.

§ 13. ACKNOWLEDGMENTS

In conclusion I wish to express my thanks to Prof. W. H. Bragg, F.R.S., for his interest in this paper, to Mr H. Bell, M.A., for reading through the manuscript and the proofs, and to the Leembruggen trust for means placed at my disposal for the construction of the instrument.

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REVIEWS OF BOOKS

Die Interferenzen von Röntgen- und Elektronenstrahlen, by M. v. Laue. Pp. vi+46. (Berlin: Julius Springer.) Price RM. 3.60.

Professor von Laue has collected in this pamphlet the chief results of the theories of the diffraction of X rays and electrons. As might be expected from anything written by him, the exposition is extremely clear, and the logical sequence of the rather complicated theories involved is explained without great mathematical detail.

In the last two chapters an account is given of the recent work on interferences in a single crystal produced when the X rays are excited in the crystal itself; and these results are compared with the corresponding effects in electron-diffraction which have been known for some time as Kikuchi's lines. Professor von Laue gives strong reasons for supposing that the latter are due to electrons which have made inelastic collisions in the crystal.

G. P. T.

The New Chemistry, by Prof. E. N. da C. Andrade, D.Sc., Ph.D., F.R.S. Pp. 58. (London: G. Bell and Sons Ltd., 1936.) 3*s*. 6*d*.

This little book which is dedicated to Professor C. D. Ellis, "who knows all about these things without believing that nothing else matters", is based on a post-graduate lecture delivered at St Mary's Hospital, London. In it the author, in his characteristically clear style, surveys recent work on the transmutation of matter which he calls "the new chemistry". The book is printed in large clear type which makes for pleasant reading, and several beautiful plates are included. The last two sentences of the book are significant and surely reflect the feelings of many physicists: "It may be that we shall never be able to utilise atomic energy, that is, mass energy, but even if this is so we may take comfort from the fact that it is certain that the atomic heat engine would be speedily followed by the atomic bomb. The pursuit of the new chemistry is none the less desirable if it does not seem possible so far to turn it to destructive ends."

H. R. L.

The Thermochemistry of the Chemical Substances, by F. R. Bichowsky and F. D. Rossini. Pp. 460, including title-page and preliminary matter. (New York: Reinhold Publishing Corporation; London: Chapman and Hall; 1936.) 35*s*. net.

Thermochemical data are based largely on the classical researches of Berthelot and Julius Thomsen, and newer researches carried out with an improved technique. The latter are dispersed throughout the literature and if a value of a thermochemical constant is required it is very difficult to find the best one. In the present monograph the authors have attempted a survey of the whole field and have given a critical set of data, in all cases explaining the reasons for their choice. The tables are very complete and include all compounds except organic substances with more than two atoms of carbon. The physical states of the substances are adequately described, and all values are recalculated in terms of moles on the basis of the 1934 international atomic weights and the calorie of 4·1850 international joules. The energy values of atoms, ions and certain molecules are given, so that the tables should also be valuable to the spectroscopist. The volume is authoritative and is a most welcome addition to the not very extensive literature on thermochemistry. It will save a great amount of time and trouble.

J. R. PARTINGTON.

Hand- und Jahrbuch der chemischen Physik, Band 9. Abschnitt III, *Anregung der Spektren*, by W. HANLE; Abschnitt IV, *Molekülspektren von Lösungen und Flüssigkeiten*, by G. SCHEIBE and W. FRÖMEL. Pp. 184+16. (Leipzig: Akademische Verlagsgesellschaft m. b. H., 1936.) M. 18.

Many of the monographs in this series, edited by A. Eucken and K. L. Wolf, have deservedly become well known to physical chemists and spectroscopists in this country. If the two now before us do not also become widely used here, the rather high price will probably be the cause. Hanle, whose monograph on the excitation of spectra occupies about three-quarters of this book, is well known for his contributions to *Ergebnisse der exakten Naturwissenschaften*, *Handbuch der Radiologie* and other collective works, as well as for his own investigations in the subject. In his present monograph, the first, and much the largest, section deals with fluorescence; the next largest with excitation by collision with electrons, atoms and ions; and the remaining shorter sections with collisions of the second kind, temperature excitation, excitation of continua, and applications to spectrographic analysis of materials and to technical light sources. The discussion includes spectra of atoms, diatomic molecules and polyatomic molecules of both inorganic and organic substances. The shorter monograph, by Scheibe and Frömel, deals in an attractive manner with the observation and interpretation of the band spectra of polyatomic molecules in liquids and solutions particularly in the visible and near ultra-violet regions. The two monographs contain, respectively, 74 and 15 figures in the text, and 17 and 9 tables of numerical data. Both are well indexed; both include numerous references to original papers, and Hanle gives also a bibliography at the end of each section.

W. J.

Klänge und Geräusche, by Dr F. TRENDELENBURG. Pp. viii+235. (Berlin: Julius Springer.) Price RM. 24.

In this exceptionally well-illustrated book the author deals with a number of aspects of sound and noise which do not find a place in other books on this subject. His connexion with the research laboratory of Siemens has given him a strong interest in the acoustics of telephony, and the book betrays this in many places. The book, in fact, is not a textbook of acoustics but forms a very interesting and readable supplementary textbook dealing with applied acoustics.

The illustrations call for praise; particularly the photographs of the structure of the ear and the X-ray photographs showing the conformation of the mouth and throat when sounding various vowel sounds.

As regards the text, one of the most important things the book teaches is the power and availability of objective acoustical measurements. As an example may be mentioned the oscillograms and analyses with which the book abounds, and which deal with a wide variety of sounds, for instance the exhaust noise of a motor cycle, speech, common musical instruments and even noise due to muscular movement. Instrumental methods are fully described in a long chapter which includes a particularly valuable account of the methods available for analysis.

An interesting account is given of the use of microphones in medicine, and oscillograms illustrating various diseases of the heart and lungs are shown. A full account is given of the effect upon music and speech of various modifications such as non-linear distortion, phase distortion and loss of certain frequency bands. The author's interest in musical matters is shown by the long chapter devoted to musical instruments, which is well supported by analyses and oscillograms. Very full attention is given to the violin, such out-of-the-way information as the form of vibration of the violin body, oscillograms of the well-known wolf tone, and even a polar diagram of the intensity-distribution round a violin being given.

Features which exemplify the up-to-date character of the book are the sections on synthetic and recorded sounds. It is surprising to discover how many systems are in use for synthesizing musical sounds and more surprising still to be told that some of them are practicable!

It is a little surprising to find that in a book which deals so fully with noise and with the acoustics of auditoria no mention is made of methods of keeping such noises from travelling into and about buildings. The omission is not a blemish, but the interest in building acoustics which the author has betrayed implies that he has information available regarding noise in buildings which could have made the book still more valuable than it is.

The above sketch of the contents of *Klänge und Geräusche* is not by any means exhaustive. Dr Trendelenburg has also dealt with the other aspects of applied acoustics which one would naturally expect to find in such a book, for instance the use of microphones and loudspeakers. The subjects which have had special mention above are those which one might not have expected to find dealt with so fully, and it is these which make the book valuable either to the advanced student who spreads his net wide or to the research worker engaged with acoustical matters.

J. E. R. C.

The Physics of Solids and Fluids, by P. P. Ewald, Th. Pöschl and L. Prandtl. Translated by J. Dougall and W. M. Deans. (Second Edition, 1936. Blackie.) 17*s.* 6*d.*

This excellent book, translated from matter prepared for the new edition of *Müller-Pouillet*, is distinguished by the way in which stress is laid on the behaviour of real substances—crystalline solids and viscous fluids—as against the isotopic solids and ideal liquids to which the books of, say, thirty and more years ago were almost exclusively devoted. It is, no doubt, owing to the lucid manner in which recent developments are exposed that a new edition has been demanded already.

There has been a substantial revision in the part of the book due to Professor Prandtl, *The Flow of Liquids and Gases*. The main novelty is the addition of a final chapter on flow with appreciable volume changes, which includes certain sections taken from earlier chapters in the former edition, but contains notable new matter on such points as flow round aerofoils when the velocity exceeds that of sound, the resistance of projectiles and cavitation. There are small changes elsewhere, such as the addition of a short treatment of two-dimensional flow. The book is an invaluable supplement to older treatises on the "properties of matter".

E. N. da C. A.

A Text-Book of Physics, by Charles A. Culver. Pp. x+816. (New York: The Macmillan Company.) 17*s.* 0*d.* net.

Professor Culver has written a textbook very suitable for students working for examinations of Intermediate Science standard. It has been his ambition to present, and he has succeeded in presenting, the material in such a way that students of the humanities will find that science has a real cultural value. The introduction of some historical material and thumb-nail biographies of eminent physicists, and the references to original papers and to textbooks giving fuller details of the various branches of the subject, are welcome innovations in a book of this standard. The author invites constructive criticism. I suggest that in the discussion of heat units, the standard thermal unit as defined in the *S.U.N. Report* 1934 (published by The Physical Society) should be stated and that specific heat should be expressed in the appropriate units and not as a ratio. Figure 116 must be redrawn. Continuous-flow electrical calorimetric methods merit more than the passing reference that one finds. Some mention should be made of a characteristic gas equation,

e.g. that of van der Waals. Refraction at a single spherical surface should be considered, if only to emphasize that most of the dioptric power of the eye is of static origin and that the eye does *not* "function as a pin-hole camera after removal of the lens". J. H. B.

Elementary Physics, by G. STEAD. Pp. xv+560. (London: J. & A. Churchill.) (5th Edition.) 12*s*. 6*d*.

It must be most gratifying for an author to find that the demand for his work is such that edition after edition is necessary. It certainly shows that the book is fulfilling a useful purpose. But this fifth edition is especially welcomed because Mr Stead has not only revised the old text, excellent in its clarity, but has introduced some hundred pages of additional matter. In doing so he has made this text-book a much more adequate guide for first-year science and medical students, to all of whom it can be confidently and warmly commended. The author adopts a sign convention for optical problems which gives the same standard formula for mirrors and lenses, but the Committee of the Physical Society on the teaching of Geometrical Optics would surely have taken exception to the sentence, "To find the power of a lens in dioptres, express its focal length in metres and turn the result upside down."

In the discussion of the laws of viscous flow the example of the delivery of water through 10 in. and 12 in. mains is unfortunate because, in general, such flows will be turbulent.

J. H. B.

Text-book of Physics, by E. GRIMSEHL, vol. v. *Physics of the Atom*, edited by R. TOMASCHEK (7th edition). Translated by L. A. WOODWARD. (London: Blackie & Son, Ltd.) Pp. xiii+474. 17*s*. 6*d*. net.

In reviewing this translation of a volume of the well-known German textbook it is advisable to begin by stating the scope of the volume, as this differs in some respects from what would be expected in an English treatise of similar title. On the side of omissions, there is no account of the fundamental work on the electron or of the gaseous discharge in any form, and while a short account is given of ionization by electrons and a fuller one of that by α particles, no mention is made of ionization by other positively charged particles or of secondary electrons released by them. For some of these the reader is referred to other volumes. On the other hand the book includes, besides the usual topics, a short account of the structure and vibrations of solids, of phosphorescence and of the internal photoelectric effect. The electrodynamics of moving media are dealt with at some length and there is a short statement of some of the problems of stellar physics. Temperature radiation is discussed, but without proofs of some of the main theorems.

The book is well produced, and the treatment is in general clear and satisfactory. A number of interesting points are mentioned which do not generally figure in a book of this character, but on the other hand the explanations are sometimes so condensed as to be difficult to follow and occasionally are even misleading, as in the case, for instance, of the account of the focusing action of the Aston mass spectrograph. In view of the admitted unreliability of his work it is unfortunate that so many of Rupp's results are mentioned, including some that are almost certainly false. In dealing with the properties of moving media Dr Tomaschek favours an ether carried by the earth, but the special theory of relativity is stated briefly. The translator has done his work well, and there are only a few clumsy sentences. G. P. T.

Principles of Electric and Magnetic Measurements, by P. Vigoureux and C. E. Webb. Pp. 392. (Blackie & Son Ltd., London and Glasgow.) 20*s*. net.

This is a textbook on experimental electricity and magnetism intended for the use of students. It differs from most previous works of a similar character in that obsolete methods of measurement are almost completely excluded, and attention is concentrated on those employed in modern research and testing laboratories, and especially methods with industrial applications. Thus part I, Electricity, includes direct and alternating current bridge methods, potentiometers, methods for radio-frequencies, thermionic valves, measurements on electrolytes, dielectrics, X rays, etc., while part II includes measurements of magnetic materials.

The proportion of the book devoted to magnetic measurements is surprisingly large. There are, for example, about 80 pages dealing with measurements of magnetic permeability and related quantities, while only 6 pages are devoted to dielectrics and 14 to electrolytes. Similarly much more space is devoted to magnetostriction than to piezo-electricity, which is surely far more important. The general student will probably find the magnetic section too detailed, though it will be very welcome to the specialist. It is to be hoped that there will be an opportunity of adjusting the balance at some time or other, for there is certainly room for a book such as this, which presents the subject from a point of view different from that of the academic writer. It will be found a useful supplement to the more orthodox works.

L. H.

Manual of Meteorology, vol. II. *Comparative Meteorology*, by Sir Napier Shaw, with the assistance of Elaine Austin. Pp. xlviii + 472. (Cambridge University Press.) 36*s*. net.

This is a revised edition of the second of the four volumes of the *Manual of Meteorology*. It may be described as a gazetteer of the meteorology of the globe, and is, in the main, an account of the ascertained facts relating to the atmosphere over different parts of the earth. Its outstanding feature is a series of monthly and annual charts of temperature, cloud-amount, rainfall, and pressure at sea level. In each case a separate chart is given for each hemisphere, the projection used showing the pole at the centre of the chart, with equally spaced parallels. These charts are extremely useful for purposes of reference, and are so well reproduced that it is easy to extract from them any material fact. It is also a novelty in meteorological textbooks to have each month of the year shown, instead of just the extreme months January and July. The intermediate months are in many ways even more interesting than the extreme ones, and the reader can here see the intermediate stages of such a transformation as that which occurs over Asia when the Siberian anticyclone of January is replaced eventually by the depression to the North-West of India in the summer. Many other facts are shown in charts, diagrams, or tables, and most of the known facts of the diurnal and annual variation of the meteorological elements and of their variation in the vertical appear in the book in one form or another.

A whole chapter is devoted to the vexed question of cycles in weather, though the author refrains from giving an opinion as to the value or reality of these.

The volume is a valuable work of reference, for it gives a mass of information not to be found elsewhere without much labour. The extent to which it is up-to-date may be gathered from the fact that on p. 421 are given diagrams of the vertical distribution of the mean winds during the month of December 1935 at three stations in the British Isles.

D. B.

Proceedings of the Plenary Meeting of the Comité Consultatif International pour les Communications Téléphoniques à Grande Distance at Budapest 1934. Vols. I–V. Pp. 660. English translation. (International Standard Electric Corporation.) 25*s.* 0*d.* net.

Although telegraphy and telephony were among the earliest applications of electricity and attracted the study of some of the most eminent physicists of the last century, interest in more modern times has moved from what is often regarded as the elaboration of classical conceptions. But the science thus handed to the communication engineer was a living study, and a consideration of present technique shows it to be based on a large body of scientific work of a high order. The present volume, the proceedings of the Comité Consultatif International pour les Communications Téléphoniques à Grande Distance (C.C.I.F.), forms the best medium for such a consideration, since it is an authoritative and comprehensive handbook of the science and practice of telephonic transmission, representing the consensus of international expert opinion. Nor is the scope limited by the title. Nearly any circuit may now serve as a link in an international or long-distance conversation, and the principles of the C.C.I. are accepted as the standard of the best telephone practice.

The interest of the physicist will, however, mainly be drawn to the possibility of his making new contributions to this branch of engineering, particularly in view of the recrudescence of interest in some of the less fundamental or less atomic or philosophic branches of physics. There are many such possibilities. The sections relating to definitions and measurements of desired sounds (speech and music), unwanted sounds (noise) and their equivalents show a lack of agreement and a measure of incoherence as to some of the basic concepts. Much essential statistical work of a general nature has been carried out on the nature of transmission and its impairment by continuous electrical disturbances, but the evaluation of discontinuous disturbance, which involves the physical nature of sound and the integration-time reaction of the ear, has received only a cursory consideration. On the other hand it can be argued that the telephone engineers have defined and largely determined means of measuring the acoustical quantities essential to the exercise of their profession; but it is very desirable that common absolute bases should be arrived at, so that results obtained in different fields of acoustical work may be readily comparable. It is believed that this question received attention at the recent C.C.I. meetings this year (1936) in London and Copenhagen. As regards Corrosion also the volume shows that work is well forward on such aspects as the distribution of the stray currents and potentials causing electrolytic damage, but not so much attention has been paid to the chemical and physical details of the electrochemical actions involved.

The main work of the C.C.I.F. and the greater part of the present volume relate to transmission, and this is studied in detail. Not only are the general types of circuits specified but also the components to be employed. In the text will be found the characteristics of equipment at present available and detailed descriptions of alternative methods of measurement and testing, while desirable directions of progress are indicated. Information on the most recent developments has its place in the volume, and it is interesting to note the extent of the study which the C.C.I.F. had already given in 1934 to the problems of the different types of transmission along lines by means of high frequency currents, and the uses to which they could be put.

Accordingly the physicist in search of new fields of practical endeavour will be well repaid for a study of the proceedings of the C.C.I.F. He will also derive much benefit from observing how basic difficulties and uncertainties can be overcome by means of limited concepts based on a careful statistical analysis, and how the complicated problems offered by elaborate circuits and equipment can be ingeniously simplified by the use of

standard forms. Grateful acknowledgment should be made to the International Standard Electric Corporation for making an English translation available. No doubt the C.C.I are wise in having an official text in one language only (diplomatic experience rather suggests that this is so) but the excellence of the present translation indicates that the difficulties, at least as regards English, might well have been overcome.

Numerical Data on Rotatory Power, by E. DARMOIS; *Numerical Data on the Raman Effect*, by M. MAGAT; *Numerical Data on Radioactivity*, by I. JOLIOT-CURIE, B. GRINBERG and R. J. WALEN. Pp. 68, 112, 57. (Paris: Gauthier-Villars.) 8*s*., 12*s*., 8*s*.

The problem of keeping in touch with the literature of physics is one which troubles most of us. It has a sub-problem, that of finding published numerical values when they are needed. The difficulty seems to have been first seen and met in Germany, where "Landolt" has ever been a help in time of trouble, but the rapid rate of growth of scientific literature, and the general upset of currencies have made so many supplementary volumes necessary, each of them at so high a price, that only the better endowed libraries can buy them without serious thought.

In France, through the International Union of Pure and Applied Chemistry, the method has been to publish volumes of data from time to time. Originally, the intention was to publish each year a single volume which would contain the data from the preceding year's literature. Unavoidable difficulties held up the work at times, and later volumes were issued containing the data for three or four years together.

The parts listed in the heading above all belong to volume XI, and it is understood that in future all volumes will be divided into a number of parts which can be purchased separately.

Two at least of the sections now under notice can fairly be classed as of topical interest, and may well expect to command a wide sale. The nuclear physics section in particular forms a very valuable compendium of the results of recent, and moderately recent, work. The section on Raman spectra differs slightly from most of the "Données Numériques" in that critical values are given, instead of the observed values. Lines observed by different authors, if agreeing within 10 $cm.^{-1}$, are assumed to be identical, and only the mean is then given.

The main difficulty in such a series of annual or quasi-annual volumes is to know in which one an isolated piece of information is to be found; to assist the user from this point of view, it is proposed to issue during the present year a substance-index covering Vols. 6–10. Such an index for Vols. 1–5 appeared some twelve years ago, and a third index volume is promised next year, to deal with Vols. 11 and 12, which will themselves contain all data up to the end of 1936.

For the sake of those who are not familiar with the series, it may be added that the main language is French, but that most explanations are given in English as well. The headings are given in addition in German and Italian. If we may venture a suggestion, it is that the size and cost of the parts might be appreciably reduced if French alone were used. A heading which prints the word "Raman" four times, accompanied in turn by Effekt, Effect, Effet and Effetto seems to be over-indulgent to the reader.

Similarly, an index of chemical names in either English or French is quite intelligible, even to a reader who has no knowledge of the language selected, provided that he knows the other. (If he knows neither, he will not be helped by having both inserted.) It may be added that full information as to past and future parts, as well as those now under review, may be obtained from the Institut de Chimie, 11 Rue Pierre Curie, Paris, V^{e}.

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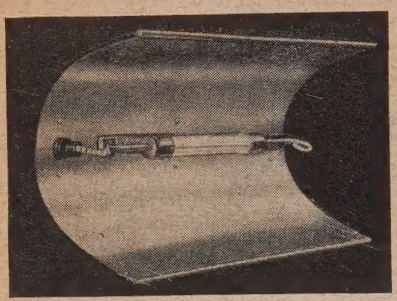

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